Chapter (1)

1.1. Introduction

Natural gemstones have for long human desire beside its economic value. They have achieved this status by possessing the desirable attributes of beauty, rarity, durability, portability, as well as the transient appeal of fashion at certain times and places. These attributes have contributed to the popularity of gemstones that has continued since ancient times.

Although new occurrences of natural gemstones are found from time to time in various parts of the world, the availability of the best quality gemstones does not always meet the current growing demand among jewellery-buying consumers. Besides the small percentage of high-quality gem material produced at a given mining locality, there is recovered a much larger percentage of lower-quality material (with poorer color and/or clarity) that has little market value. Therefore, researchers strive to develop methods to treat lower-quality gem materials to enhance their appearance and hence their saleability in the jewellery market place.

Precious stones suitable for jewellery are seldom encountered in nature; however, there are great resources of colorless types of these stones which are of little value. Therefore, with respect to the growing demands for jewellery, it is necessary to study different methods for stone enhancement [(1)] or the color of the raw stones. The color of gemstone produced or altered by irradiation is caused by radiation-induced point defects called color centers which generally take the form of vacancies.

Treated gemstones such as topaz, beryl, emerald and quartz are very common and they cost only a fraction of the cost of the natural stones. Topaz is the gemstone most often irradiated in a reactor; due to its relative purity compared to

other gemstones [2]. Reactor irradiated topaz is called "London blue" in the jewellery trade [3].

A gemstone is prized according to its beauty or perfection. Hence, appearance is almost the most important attribute of gemstones. Their beauty must stand for a very long time. Color is the main factor that makes a stone beautiful or desirable. Colors in treated gemstones by gamma irradiation are affected by heating to high temperatures, while artificial gemstones which produced by irradiation with neutrons have a stable color under effective factors such as heat. A variety of treated gem materials are encountered today in the jewellery marketplace in increasing quantities.

1.2. Color

Color is the most important characteristic of gems. Gemstones are colored by the light, their ability to absorb one color from the six that comprise white light: red, orange, yellow, green, blue, or violet. If the stone did not absorb color, it will appear colorless. If all the colors are absorbed, the stone will be black. If green is absorbed into the stone, the stone will show the complementary color to the one absorbed, in this case red. The distance that the light ray travels through the stone can influence the absorption and thus the color. The refractive index (RI) is of prime importance to the faceter. When the light passes from air to another medium like a gemstone, the light path is modified because the velocity of light is lowered in the denser medium. In other words, the light is refracted.

The amount of refraction is called the refractive index (RI) and is defined as: the ratio of the velocity of light in air to the velocity of light in the gemstone while dispersion is the ability of a gemstone to split the light into its spectral colors. This color dispersion produces beautiful plays of color, also called fire. The body color of gemstone also tends to mask dispersion if the tone is medium to dark. Some minerals such as beryl and tourmaline are able to split the light into

two separate rays which are polarized at right angles to each other. This effect is called birefringence, anisotropic or doubly refractive.

1.3. Topaz

Topaz is an aluminium silicate mineral with variable amounts of F and OH groups and its composition is $Al_2SiO_4(F,OH)_2$. It has trace impurities that vary from location to location. These impurities can affect color and activation products. The main gem topaz deposits are localized in Brazil and Russia. Other locations include Mexico, U.S.A (Texas and Colorado) Pakistan and Nigeria. Unfortunately, topaz is very rare in Egypt. Pure topaz is colorless or pale yellow. Blue topaz can occur naturally, but is usually produced via irradiation [(4)]. Topaz has a hardness of 8, a specific gravity of 3.4–3.6, and a vitreous luster. Pure topaz is transparent but is usually tinted by impurities. When this colorless topaz is irradiated and (in most cases) subsequently heated, however, an attractive blue stone may be produced [(5)]. Topaz is currently the most commonly irradiated gemstone (to get various shades and tones of blue) [(6)].

1.4. Treatments

Gems are frequently subjected to various treatments in order to improve their appearance in terms of color and transparency, and hence increase their commercial value [(7)]. By time, the irradiation process becomes cheaper and the irradiation quality is enhanced [(8)]. Color is of course a most important property of gemstones. Gemstones are frequently treated to alter their color to good or excellent results [(9)]. Usually by different techniques such as dyeing, heating and irradiation treatment; by gamma using cobalt-60 or inside cyclotrons and recently by neutrons inside research reactors, the color of the gem after irradiation is poor, good and or excellent depends on the stone and its origin [table (1) 9].

The use of radiation to treat gemstones begins early but the commercial use is recently. Amount of treated gemstones varieties now are very hug. The primary

key to the value of gemstones lies in their beauty [10]. The fashioning, sorting, marketing and selling of treated gemstones and jewellery are a large multibillion dollar industry, employing people from all over the world [11]. The treatment has a significant effect on the value of a gemstone [12]. At present, neutron and gamma radiation from nuclear reactors, gamma radiation from high-power ^{60}Co sources, and charged elementary particles from accelerators are widely used for coloring and color enhancement of natural precious stones. Irradiation induces radionuclides with different half-lives, resulting to activity. Therefore, depending on their activity and half-lives, some time is required for the radioactivity level of stones to fall to a value permissible for application in jewellery. Gamma irradiation does not result in radionuclide production [1]. The most widely used treatment for gems is heating and irradiation depending on the kind of gem and the desired effect.

Some gemstones receive heat treatment after irradiation to improve the appearance and stabilize the color by annealing out the less appealing and weaker color centers [2]. Raw samples have a very low commercial value, because it is colorless or present only weak coloration. Topaz is the best example for enhancement of color by commercial application of neutron irradiation.

1.5. Irradiation Methods

1.5.1. Gamma Irradiation

Most published reports of irradiated topaz involve the use of gamma facilities. Nassau and Prescott (1975) [13] used ^{60}Co gamma rays to deliver a total dose of 10 Mrads at a dose rate of 0.7 Mrads/hr to 86 colorless topaz samples. The color that was produced was not uniform. Nassau (1985) [5] recommended dose rates of less than 5 Mrads/hr to avoid an excessive buildup of temperature inside topaz during gamma irradiation, depending on the size of the topaz.

1.5.2. Neutron Irradiation

The blue color produced by neutron bombardment of topaz is usually uniformly deep, regardless of the size of the topaz, because neutrons have excellent penetrating properties. Unfortunately, radioactive by-products are produced with such treatment. Several studies suggested that thermal neutrons (less than 0.025 eV) create the majority of radioactive isotopes in topaz, but fast neutrons (greater than several MeV) produce the blue color [(14)]. The depth of color was not correlated with dose [(9)]. Neutron irradiation produces a deeper blue than irradiation by gammas or electrons because neutrons are more heavily ionizing, inducing more change to the crystal structure, and penetrate the whole crystal (unlike electrons) [(4)].

The difference between neutron and γ-ray irradiation appears in three factors. At irradiation time; for γ-ray may take around 9 months while in neutron irradiation case the time is in the range of hours only. The Color for γ-ray is Sky blue and Swiss blue in neutron irradiation. The gemstones gamma irradiation process to enhance the color is widely accepted for the jewellery industry [(8)].The third factor is induced radioactivity which is absent in γ-ray irradiation but it is very strong in neutron irradiation. Two types of radiation sources were used in the present study for the irradiation of the samples of treated gemstones; neutrons and gamma rays. Each one of them has some advantages and drawbacks. Irradiation with fast neutrons, or gamma rays from Co-60, induces homogeneous coloration; however the 1.17 and 1.33 MeV gamma rays from Co-60 have an extremely low efficiency in defect production [(15)].

Samples were irradiated using transition pneumatic system (quick irradiation) for short time in the range (10-30) second and long irradiation in the hours range.

The gemstone irradiation market may grow with lower irradiation prices and better color enhancement results. A fundamental aspect to be considered for a future potential economic feasibility of colored topaz induced by neutrons is the

time needed for the irradiated gemstones to decay. Radioactivity from some long-life elemental impurities would require long-time storage of the gems before being delivered.

Most current gemmological research is directed at the identification of these important gemstones. However, extensive laboratory testing may not be possible for other gems which are bought and sold in large quantity and whose individual value may be less significant. In such instances, testing for treated samples may be carried out only on representative samples in a group.

1.6. Aim of the work

1- Studying characteristics of irradiated topaz by neutrons and gamma.

2- Studying the crystal defects of raw and irradiated topaz by γ with Positron Annihilation Spectroscopy (PAS) technique.

3- Spectrophotometer measurements of topaz at different wavelengths.

4- Studying residual radioactivity in irradiated samples, aiming to reach the safety level of transportation.

5- Studying trace and ultra trace elements in used samples, by trace element analytical techniques such as Laser Ablation Inductively Coupled Plasma Mass spectrometer (LA-ICPMS).

6- Studying all samples by Raman spectrometer (RS). Samples were subjected to Raman spectroscopic studies before and after treatment.

7- Measuring irradiated samples by different radiation protection devices.

8- Analyse irradiated samples by Neutron Activation Analysis (NAA) in two cases; short and long irradiation.

Chapter (2)

Theoretical Considerations

2.1. Gemstones

There are almost 4000 known minerals, of which only about 50 are commonly used as gemstones. Those that form crystals of sufficient size and quality to be cut and fashioned as gems are referred to as 'gem quality' or 'cuttable' pieces; other minerals or rocks with particularly attractive features (color, texture, or pattern) may be called 'decorative' pieces. Crystals are usually faceted (cut and polished) to give a gemstone with a number of flat faces, while decorative stones are mainly tumbled or polished to produce pieces for personal adornment [16]. Much gemstone mining is carried out in remote places, with secrecy and security to protect the interests (and sometimes the lives) of the owners [16].

Besides natural gemstones, an extensive variety of treated gemstones are available today in the marketplace, and this trend will continue. The latter offer consumers a wider choice of gemstones with different colors, qualities and prices. These gemstones are the products of conventional and more recently developed treatment methods [17].

In general, the most widespread treatment methods, especially for colored gemstones, involve a change to the desired color. Two techniques in this category – exposure to heat and / or radiation – are similar to natural processes that can affect gem minerals in the earth; as a result, these treatments can sometimes produce similar colors as one could encounter in natural gemstones [18]. The earliest method of irradiating diamonds by highly energetic particles is known since the late 19th and early 20th centuries, when Antoine-Henri Becquerel and after him Sir William Crookes experimented on the effect of radium on diamond. They rendered the diamonds green by subjecting them to

the alpha particles emitted by the radium. However, this process induces long-lived radioactivity.

The first report of the production of blue topaz after Irradiation was by F. H. Pough and T.H. Rogers in 1947 [19]. Since then, there have been various reports on topaz irradiation using an assortment of treatments, such as gamma rays, neutrons, and electrons. The production of blue topaz from colorless one by irradiation was first reported by F. H. Pough in 1957 [20] as one of a large number of color changes observed in a variety of materials subjected to such treatment. As early as in the year 1974, K. Nassau [21] reported that much colorless gem material can be turned into yellow-brown color on irradiation, such as quartz and topaz. Unfortunately, these reports give little, if any, information on the radionuclides, or the levels of radioactivity produced during irradiation, or the type of detection equipment necessary to ensure that radiation levels are within acceptable limits [9]. Later, irradiation with gamma rays by exposure to cobalt-60, with protons and deuterons via a cyclotron, with neutrons in a nuclear reactor and with electrons by electron accelerators replaced the dangerous radium treatment; of these, electrons and neutrons are the most common particles used today for diamond treatment. These irradiations do not create residual radioactivity as long as they are properly applied. If not properly performed, radionuclides can be created, rendering them radioactive. Neutrons especially are known to strongly activate materials when they are used with out caution [14].

The blue topaz industry has grown dramatically through years and now accounts for a $675 million in the worldwide market [9]. It is the one of the highlights in international gem trading. It is estimated that the market absorbs about 100 million carats per year, and today nearly 100% of such topaz is treated by laboratory irradiation [3].

In gem minerals, there are two different color mechanisms. One is related to optical transitions involving transition metals, including also charge transfer

transitions between two of them or its oxygen neighbors. The other cause of color is related to intrinsic color centers. Two types of centers, electron and hole centers, commonly produce color by irradiation. A well-known example is the smoky color of quartz. It is easily created in colorless quartz that contains silicon replacing Al impurities to a large amount [3].

2.2. Gemstones Treatments

The treatment of gemstones by various means to improve their appearance extends in some cases back to thousands of years. Prior to modern times, the availability of treated gem materials was restricted, and knowledge of them was not widely known. The development of the science of mineralogy was started in the 1800s. The past century witnessed growth of gem treatment in terms of new techniques utilized on a widening range of gem materials [17].

The treatment creates special care requirements for the gemstone to retain the benefit of the treatment; and it has a significant effect on the value of the gemstone [12]. Enhancement is defined as any traditional process other than cutting and polishing that improves the appearance (color/clarity/phenomena), durability or availability of a gemstone. A gemstone enhancement is considered permanent as long as the effect of the enhancement does not change under normal wear, cutting, repair, cleaning or display conditions.

Gemstones have historically and traditionally been enhanced. The methods of the enhancement processes vary within each variety and change as new and better methods are developed [12].

All natural gemstones can be divided into three basic categories:

a) Those which are not enhanced (N).

b) Those which are normally enhanced (E).

c) Those treatment processes not covered under the “N” or “E” symbols are addressed in a specific manner [12].

Sometimes multiple enhancement techniques are applied to the same material. All treatments must be listed at the stone certificate.

Mineral color is a result of physical structure and/or the interactions of light with electric fields in a mineral. Gemstones are generally harder and heavier than the surrounding minerals [16]. Many gemstones are not normally an attractive color. Exposing them to radiation turns it from a clear or light straw color to being nicely blue [22].

Exposure to ionizing radiation can alter a mineral's distribution of electric charge. Exposure to particle radiation can also change a crystal's physical structure. Impurities can also affect color by stressing the crystal structure and influencing electron configuration. The variety of inclusions in gemstones and their use in identification are described in numerous articles and books, including the reference works by Gübelin and Koivula (1986, 2005, 2008), which contain thousands of photographs [23].

2.2.1 Treatment methods

2.2.1.1. Coating

A gemstone having a body with a pavilion bearing a coating that serves as an absorber of certain visible radiation frequencies to provide color so as to impart in the gemstone a desired uniform body color that does not substantially change in hue when viewed at different angles of observation. The use of such surface enhancements as lacquering, enameling, inking, foiling to improve appearance, provide color or add other special effects. The coat layer is high absorption.

2.2.1.2. Diffusion

Diffusion treatment is a surface layer baked into the gemstone surface that imparts a false color for the stone. Diffusion will bake the coloring element into the surface of the stone to a minimal depth. It is not permanent. It will show through with scratches from everyday wear and tear. So while they are cheap,

they are also temporary. So, any sale or purchase of these stones should be accompanied by a full disclosure of the process at the time of sale.

2.2.1.3. Heat Treatment

The heat treating to enhance, clarify or create color in a stone is the most common [24]. Basically, it rearranges the atoms in the gemstone. The stone is heated to a very high temperature, which causes the inclusions to reform themselves and improve the color by making darker, lighter, more intense, or of a different color. Any type of furnace can be used, and heating in air is perfectly adequate. After the required time at temperature, the furnace is often cooled at an equally slow rate and the stones are not removed until they have returned to room temperature [5]. Sometimes re-crystallization occurs on the surface of these stones at the higher temperatures so that they must be re-polished [25].

2.2.1.4. Irradiation

Irradiation is the process by which gemstones are exposed to various types of nuclear radiation to change some parts of the crystal structure. This change in crystal structure causes the gemstone to alter its color, either by the addition or subtraction of some part of the crystal structure. Basically, the radiation bombards the atoms in the gemstones, knocking them either out of the stone, or moving them around inside the stone. The removal from the stone, or relocation inside it, causes the stone to react to light differently, which causes the stone to offer a different color.

2.2.1.5. Fracture Filling

Fracture filling is often referred to as "Clarity Enhancement". It is the filling of surface breaking fissures in the stone. The process is not completely permanent, but does provide some nice looking stones.

2.2.1.6. Laser Drilling

Laser drilling is the process by which high powered lasers are used to burn out inclusions in a stone. These lasers burn the inclusion from inside out, making a hole that goes from deep inside a stone to the surface.

2.2.1.7. Oiling

Oiling/resin infusion is the filling of surface-breaking fissures with colorless oil, wax, resin or other colorless substances, except glass or plastic, to improve the gemstone's appearance.

2.2.1.8. Bleaching

The use of heat, light and/or other agents to lighten or remove a gemstone's color is called bleaching.

2.2.1.9. Dyeing

Is the introduction of coloring matter into a gemstone to give it new color, intensify present color or improve color uniformity. The stability of dyed gems is dependent upon the type of the dye.

2.2.1.10. Quench crackling

If the gem is not porous or fractured naturally, the opening for the dye to enter the stone is produced by "quench crackling," a heat-induced thermal shock that creates a network of fractures (Hurlbut and Kammerling, 1991) [26].

2.2.1.11. Impregnation

The impregnation of a porous gemstone with a colorless agent (usually plastic) is used to improve durability and appearance.

Gems treated by heating and irradiation are most common while the other methods such as surface [coating or diffusion] and dyeing are less frequently encountered in the marketplace [17].

Nowadays, the preferred treatment methods are primarily electron irradiation with still some neutron irradiation; gamma irradiation [27]. Stones are colored when some process removes some wavelengths (absorbs specific wavelengths) from the visible spectrum.

2.3. Topaz

It is the famous treated gemstone at the market [28]. The blue color in topaz is caused by color centers, while impurities can also cause colors as in the following:

Table (2.1); some impurities and their effects [4]

Color	Impurity
Red/Pink	Chromium
Blue	Titanium & Iron
Yellow	Iron & Unknown
Orange	Chromium & Iron
Violet/Purple	Chromium & Titanium and/or Iron
Green	Iron

2.3.1. Color at topaz

Topaz has many different colors all of it due to color centre except for the pink-to-violet and the pink component are caused by a chromium impurity [29].
The most common colors of topaz resulting from radiation treatments are:
Sky Blue, London Blue and the Swiss Blue color. A color center involves one electron missing from a normally occupied position, leading to a 'hole color-center', while the presence of one extra electron leads to an 'electron color-center' [30]. Color establish also from band gaps in semi conducting minerals often result in intense colors. Inclusions of foreign phases are often responsible for the color of minerals.

2.3.2. Process of cooling

Because of the high power deposited in the stones during the irradiation processing, it is important to cool the stones. Water is used for the cooling and the stones are either placed in running water during the processing.

2.3.3. Radioactivity in gemstones

Radioactivity in gemstones resulted from γ facilities is very weak while gemstones treated with neutrons in a nuclear reactor turn radioactive for a longer time, usually from a few months up to years.

Theoretically, a few weeks after neutron irradiation in a reactor, chemically pure topaz would not be radioactive because the neutron-activated major constituents of topaz (fluorine, aluminum, oxygen and silicon) have half-lives of only seconds to hours [28].

2.4. Nuclear reactor

Nuclear reactor is a device for producing controlled release of nuclear energy. Reactors can be used for research or for power production. A research reactor is designed to produce various beams of radiation for experimental application; the heat produced is a waste product and is dissipated as efficiently as possible.

2.4.1. Self-Shielding

In some locations within the reactor, the flux level may be significantly lower than in other areas due to a phenomenon referred to as neutron shadowing or self-shielding. Because of it the wrong choice for irradiation box makes the half sizes of stones colored and the other still without any changes.

2.5. Neutron Activation Analysis (NAA)

This study considers the equipment and the analytical method of gamma-ray spectroscopy by illustrating its use to determine the type and quantity of residual radioactive nuclides present in the laboratory irradiated gemstones. The analysis also revealed the treatment processes used and the dates of decay of the radionuclides in the gemstones [31].

NAA has played an important role in science and technology [28]. It is sensitive for most elements in the periodic table with good accuracy and

precision, non-destructiveness and multi-elemental analysis ability. NAA has become an essential method in the trace elemental analysis.

2.5.1. Gamma-Ray Emission

One or more gamma photons can be emitted from the excited states of daughter nuclei following radioactive decay. A gamma-ray spectrum is characteristic of the particular radionuclides that are present. By techniques of gamma-ray spectroscopy the intensities of photons at various energies can be measured to determine the distribution of radionuclides in a sample. When ^{60}Co is present, for example, photons of energy 1.173 MeV and 1.332 MeV are observed with equal frequency.

2.5.2. Principle

The process for analyzing samples by NAA involves irradiating them with a neutron source. Neutrons are captured by elements in the sample to produce radio-nuclides. Gamma-rays are emitted from the radio-nuclides as they decay. The energies of these gamma-rays are, in general, distinct for a specific nuclide and the rate at these photons are emitted with a particular energy can be measured using high-resolution semiconductor detectors. Because the production and decay rate of gamma-ray radiation are dependent on the half-life of the nuclide, elemental measurements can be optimized by varying the irradiation and the decay times. A half-life of a radioactive material is the time it takes one-half of the atoms of the radioisotope to decay by emitting radiation. The half-life of a radioisotope can range from fractions of a second (radon-220) to millions of years (thorium-232) [32].

There are, at present, only two detectors categories of major importance: inorganic scintillators, of which NaI(Tl) is the most popular, and the germanium semiconductor detectors [33]. Germanium detector is preferred for the analysis of complex gamma-ray spectra involving many photon peaks [33].

2.5.3. Procedure

The most common procedure for NAA involves encapsulating the samples and suitable standards in heat-sealed polyethylene or quartz vials and simultaneously irradiating them. Ideally, the samples and standards have to experience the same neutron fluence. Following sequential decay periods, each standard sample is counted utilizing high resolution germanium detectors coupled to a multi-channel analyzer system. The multichannel analyzer (MCA) is operating on the principle of converting an analog signal (the pulse amplitude) to an equivalent digital number. The analog to converter (ADC) is a key element in determining the performance characteristics of the analyzer [(33)]. Such counting of samples over sequential decay periods optimizes the determination of 35 to 50 elements in various types of samples. The analyzer system converts the signals that result from gamma-ray photons impinging the detector into digital electronic pulses. Gamma-ray counts accumulated in an energy region above the background counts produce photon peaks. After counting is complete, these data are processed using computer programs that smooth the spectral data and determine the net areas of gamma-ray photon peaks and translates the area into count rates (counts per minute or cpm). These programs are capable of resolving overlapping and complex photon peak energy regions. Additional data for decay time differences, electronic dead time losses and unresolved interferences and compares the sample data (cpm/weight) to the standard data (cpm/μg) to calculate elemental abundance in the sample.

2.5.4. Background in gamma-ray spectra

Cosmic radiation, radioactive contamination of shielding material, radio activity within the detector and unidentified sources are the most contribution. The background in gamma-ray detectors increases roughly as detector volume. Therefore it is important to select a detector size that is not longer than necessary to give a reasonable counting efficiency for the samples counting [(33)].

Although the background is expected to be nearly steady state in time, it may show a perceptible variation over periods of hour or days.

Some additional interfering radiation can be observed as a result of the interaction of primary gamma rays from the source with structural and shielding material around the detector [33].

2.5.5. Radiation Measuring Devices

Measuring device or instrument is that shows the extent or amount or quantity or degree of something. In the physical sciences measurement is the activity of obtaining and comparing physical quantities of real-world objects and events. Established standard objects and events are used as units, and the measurement results in a given number for the relationship between the item under study and the referenced unit of measurement. Measuring instruments or devices are the means by which this translation is made. All measuring instruments are subject to varying degrees of instrument error and measurement uncertainty.

At most radiation measuring devices the reverse-biased n–p junction constitutes an attractive radiation detector. The depletion region has high resistivity, and ions produced there by radiation can be collected swiftly and efficiently. It can serve as a rate meter or to analyze pulses. The number of electron–hole pairs produced in a pulse is proportional to the energy absorbed in the active volume, and so the junction can be used as a spectrometer [34].

2.5.5.1 Errors in Analysis

These errors in the analysis by NAA should be reduced as possible [28].

Figure (2.1); examples of HPGe detectors (Canberra)

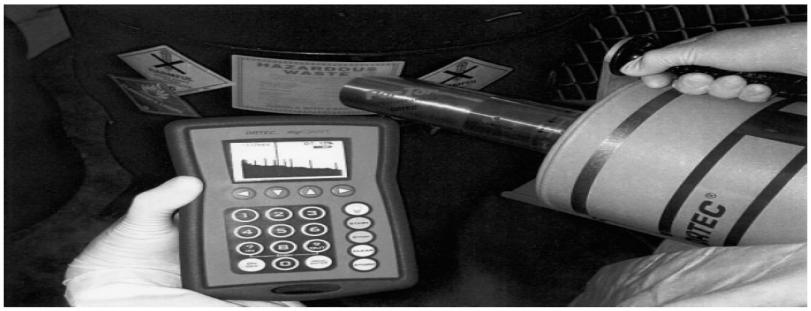

Figure (2.2); Portable HPGe multichannel analyzer(ORTEC)

2.5.6. Detection Limits

Elemental detection limits for NAA are variable because the production of radioactive nuclides depends on the cross section of a specific element. Other factors should be considered [28].

2.5.7. Concentrations calculation methods

2.5.7.1. Relative Method

Sample and element standards are irradiated simultaneously and later measured under the same counting conditions. The concentration of the element(s) of interest is calculated by comparison of the measured activity between the sample and the standard.

2.5.7.2. Non-Relative Methods

2.5.7.2.1. Absolute method

Sample and flux monitors are irradiated simultaneously and measured under the same conditions. The neutron flux can be measured from the activity of the flux monitors and can be applied in the neutron activation equation where the concentration of the elements under consideration can be obtained directly. The absolute method suffers from several significant difficulties. These include uncertainty in nuclear decay constants, and inability to measure accurately absolute irradiation parameters.

2.5.8. Neutron Irradiation of Gemstones

As a result of neutron irradiation to elements of the gemstones, radioactivity is produces. The activity of a pure radionuclide decreases exponentially with time. If N represents the number of atoms of a radionuclide in a sample at any given time, then the change "dN" in the number during a short time "dt" is proportional to N and to dt. Letting λ be the constant of proportionality, we write:

$$dN = -\lambda N dt \rightarrow \quad (2.1)$$

The negative sign is needed because N decreases as the time t increases. The quantity λ is called the decay, or transformation, constant; it has the dimensions of inverse time (e.g., s^{-1}). The decay rate, or activity, A, is given by:

$$A = -dN/dt = \lambda N \rightarrow \quad (2.2)$$

We separate the variables in Eq. (2.2) by writing

$$dN/N = -\lambda\, dt \rightarrow \quad (2.3)$$

Integration of both sides gives

$$\ln N = -\lambda t + c \rightarrow \quad (2.4)$$

Where "C" an arbitrary constant of integration and is fixed by the initial conditions. If we specify that N_0 atoms of the radionuclide are present at time t = 0, then Eq. (4.4) implies that $c = \ln N_0$. In place of (4.4) we write

$\ln N = -\lambda t + \ln N_0 \rightarrow$ (2.5)

$\ln(N/N_0) = -\lambda t$ (4.6) or $N/N_0 = e^{-\lambda t} \rightarrow$ (2.6)

Equation (2.6) describes the exponential radioactive decay law. Since the activity of a sample and the number of atoms present are proportional, activity follows the same rate of decrease,

$A/A_0 = e^{-\lambda t} \rightarrow$ (2.7)

Where A_0 is the activity at time t = 0. The dose rate at a given location in the neighborhood of a fixed radionuclide source also falls off at the same exponential rate.

During successive times T, called the half-life of the radionuclide, the activity drops by factors of one-half.

High-purity germanium (HPGe) detectors are available for measurements of radioactivity. The germanium crystal of the spectrometer is housed in a vacuum-tight cryostat unit, which typically contains the preamplifier in a cylindrical package. Depending on the intended application, germanium detectors come in a number of different planar and coaxial configurations. Nuclide Identification in Gemstones and activity calculations are performed by using stored calibration information. The instrument can also interface with a computer to utilize other software applications [34].

2. 6. Inductively Coupled Plasma Mass Spectrometer (ICP-MS)

Even if the rule is that gem analyses must be nondestructive, it might be necessary in some cases to resort to micro destructive techniques, some of which are becoming increasingly popular in gemology laboratories. The main micro destructive techniques in gemology are laser-induced plasma and laser ablation–inductively coupled plasma–mass spectrometry (LA–ICP–MS). Both techniques allow the analysis of trace elements with detection limits down to the ppm level (or lower) and does not require any preparation of the samples [(23)]. For LA-ICP-MS analysis, the absolute amount of materials ablated during each run varies from one to another due to differences in the sample matrix and the related absorption behavior of the wavelength used for ablation (e.g., Hemmerlin and Mermet, 1996; Horn et al., 2001). An internal standard is generally used to correct the variations in the absolute amount of materials ablated during each run (Longerich et al., 1996). In other words, the concentration of at least one element must be known or determined by an independent method prior to LA-ICP-MS analysis. The elements used as internal standards (e.g., Si, Mg, Ca) can vary widely [(35)].

The sensitivity achievable ICP-MS for any element depends upon its isotopic abundances, its degree of ionization in the ICP, the ion lens voltage, and parameters associated with sample introduction and plasma operation[(28)]. The inductively coupled plasma-mass spectrometer permits novel analysis of trace elements in complex minerals [(28)].The Inductively Coupled Plasma is formed within the confines of three concentric glass tubes or plasma torch. Each tube has an entry point, with those of intermediate (plasma) and external (coolant) tubes being arranged tangentially to that of the inner tube, where the later consists of a capillary tube through which the aerosol is introduced from the nebulization / spray chamber. A coil of copper tubing through which water is recirculated is located around the outer glass tube. The input power to the ICP is achieved through this load or induction coil, and is typically in the range of 0.5-

2 kW at a frequency of 27 or 40 MHz. The input power induces an oscillating magnetic field, whose lines of force axially orientated inside the plasma torch and follow elliptical paths outside the induction coil. At this point, no plasma exists. In order to initiate plasma, the carrier gas flow is first switched off and a spark is then provided momentarily from a Tesla coil, which is attached to the outside of the plasma torch by means of a piece of copper wire. Instantaneously, the spark, which is a source of 'seed' electrons, causes ionization of the argon carrier gas. This process is self sustaining, so that argon, argon ions and electrons now co-exist within the confines of the plasma torch, and can be seen protruding from the top in the shape of a bright white luminous bullet. This characteristic bullet shape is formed by the escaping high-velocity argon gas causing the entrainment of air back towards the plasma torch itself. In order to introduce the sample aerosol into the confines of the hot plasma gas (7000-10000 K) the carrier gas is introduced; this 'punches' a hole in the center of the plasma, thus creating the characteristic doughnut or toroidal shape of the ICP. Laser ablation combined with high resolution inductively coupled plasma mass spectrometry provides a new approach to the analysis of quartz at the ng/g level [36].

2.6.1. Plasma Sources

At the ICP, there is inert gas (argon) plasma maintained by the induction of a radio-frequency (RF) field and ionized argon gas which flows through the quartz torch. It appears as a very intense, brilliant white, teardrop-shaped discharge. RF power (in kw) is applied to a Cu load coil and the induced oscillating electric and magnetic fields to be set up in the area at the top of the torch. The different plasma zones are:

- Preheating zone in which desolvation, vaporization and atomization of the sample takes place.

- Induction zone in which the inductive energy transfers from the load coil to the plasma takes place (T=10 000K).
- Initial radiation zone is for excitation and ionization (T=8000 K).
- Normal analytical zone in which analyte emission typically measured (T=5000 to 8000K).
- The temperature of plasma tail is less than 5000 K [28].

The LA- ICP-MS method reported in this study requires minimal preparation of the sample, reduces the possibility of contamination, and is rapid and cost-efficient. The trace element distribution in quartz opens a rarely used source of primary information that may be applicable to a wide variety of geologic problems, including igneous geochemistry, provenance studies and rapid feasibility studies of industrial quartz resources [36].

2.7. Raman Spectroscopy

Raman spectroscopy is an ideal method for the examination of marketable gemstones because of the lack of sample preparation involved and the nondestructive nature of Raman analysis.

Raman Spectroscopy (FTIR) is a powerful tool for identifying types of chemical bonds in a molecule by producing an infrared absorption spectrum that is like a molecular "fingerprint". This means that Raman spectroscopy is a useful tool in the molecular characterization of inorganic species. This vibrational technique has certain advantages over the other techniques such as the small quantity of sample required, quick and easy sample preparation and short analysis time [37].

Raman spectroscopy provides a method for the examination of real and treated gemstones [6]. It can determine the nature of inclusions deep inside gemstones. As most gems are inorganic, the presence of organic molecules can be proof of impregnation with a resin, oil, or polymer.

Raman spectroscopy comprises the family of spectral measurements made on molecular media based on inelastic scattering of monochromatic radiation. During this process energy is exchanged between the photon and the molecule such that the scattered photon is of higher or lower energy than the incident photon. The difference in energy is made up by a change in the rotational and vibrational energy of the molecule and gives information on its energy levels; hence Raman spectroscopy could provide information about molecular structure.

When a beam of light is impinged upon a sample, photons are absorbed by the material and scattered. The vast majority of these scattered photons has exactly the same wavelength as the incident photons and is known as Rayleigh scatter, but a tiny portion (approximately 1 in 107) of the scattered radiation is shifted to a different wavelength. These wavelength shifted photons are called Raman scatter. Most of the Raman scattered photons are shifted to longer

wavelengths (Stokes shift), but a small portion are shifted to shorter wavelengths (anti-Stokes shift) [28].

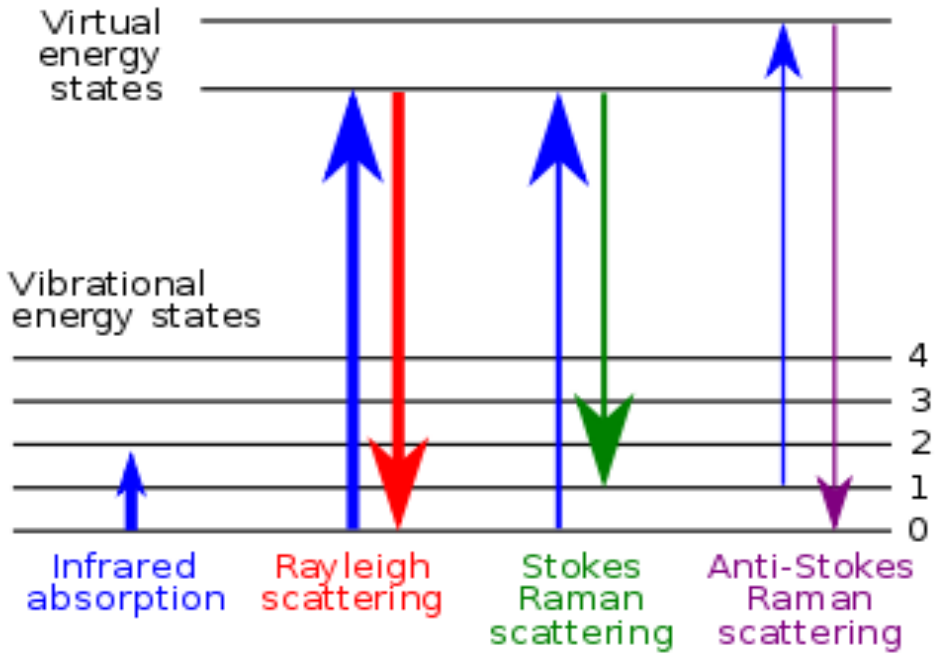

Figure (2.3); Energy level diagram showing the states involved in Raman signal. The line thickness is roughly proportional to the signal strength from the different transitions.

In each case, the incident photon excites an electron into a higher virtual energy level and then the electron decays back to a lower level, emitting a scattered photon. In Rayleigh scattering the electron decays back to the same level from which it started. In both types of Raman scattering the electron decays to a different level than that where it started. Stokes Raman scattering occurs when the final energy level is higher than the initial level, while anti-Stokes Raman scattering occurs when the final energy level is lower than the starting level. Stokes scattering is much more common than anti-Stokes scattering because at any given time an electron in the most common temperature range is most likely to be in its lowest energy state, in accordance

with the Boltzmann distribution. Only Stokes Raman scattering is commonly used in spectroscopy.

2.7.1. Raman Instrumentation

The typical Raman spectrometer consists of three basic parts; the laser, the collection device and the spectrograph [28]. Laser is used to excite Raman spectra because it gives a coherent beam of monochromatic light. This gives sufficient intensity to produce a useful amount of Raman scatter and allows for clean spectra, free of extraneous bands. Lasers used for Raman spectroscopy must exhibit good wavelength stability and low background emission.

The probe is a collection device that collects the scattered photons, filters out the Rayleigh scatter and any background signal from the fiber optic cables, and sends the Raman scatter to the spectrograph. Many probes also focus and deliver the incident laser beam.

Raman can be used to rapidly distinguish various gemstones that do not readily lend themselves to identification by other, more traditional gemological identification methods. Raman spectra also can provide a unique fingerprint of the gemstone with the ability to distinguish any future alteration of the gemstone. The photoluminescence of colored diamonds offers a method for the determination of the color of diamonds and provide a further discrimination of the diamond color scale [28]. Activation energy could be calculated to distinguish between the effect of gamma rays and neutron interaction with the topaz stone.

2.8. Positron annihilation spectroscopy (PAS)

2.8.1. Introduction

The positron is the antiparticle or the antimatter counterpart of the electron. The positron has an electric charge of +1, a spin of 1/2, and the same mass as an electron. When a low-energy positron collides with a low-energy electron, annihilation occurs, resulting in the production of two gamma ray photons. The first scientist deemed to have captured positrons through electron-positron annihilation was Chung-Yao Chao, a graduate student at Caltech in 1930, though he did not realize what they were at that time.

Positrons may be generated by positron emission radioactive decay (a weak interaction), or by pair production from a sufficiently energetic photon. The existence of positrons was first postulated in 1928 by Paul Dirac as a consequence of the Dirac equation. In 1932, positrons were discovered by Carl D. Anderson, who gave the positron its name [38].

Today, positrons are routinely produced in positron emission tomography (PET) scanners used in hospitals and in accelerator physics laboratories used in electron-positron collider experiments. In the case of PET scanners, positrons provide a mechanism to show areas of activity within the human brain.

Annihilation γ-ray is equal to 0.511 MeV which is named death ray. It is produced from positron annihilation in the used material. The positron emission from ^{22}Na isotope is simultaneous with the emission of a 1.28 MeV, which can be used as a birth gamma. Thus the lifetime of the positron is the time delay between the birth and annihilation gamma rays.

Positron annihilation spectroscopy (PAS) or Positron lifetime spectroscopy is a non-destructive spectroscopy technique to study voids and defects in solids. It is a result of an encounter of the electron with its antiparticle - Positron. The energy released by the annihilation forms two highly energetic gamma photons, which travel in opposite direction. These gamma rays provide a useful analysis tool which has found many practical applications in physics, chemistry and

medicine. There are three methods of this technique; positron lifetime spectroscopy, Doppler broadening of the annihilation line and the angular correlation of annihilation γ-quanta.

2.8.2. Positron Decay (β^+)

Some nuclei, such as $^{22}Na_{11}$, disintegrate by emitting a positively charged electron and a neutrino. Positron decay has the same net effect as electron capture, reducing The atomic number, Z, by one unit and leaving the mass number, A, unchanged. Thus the mass of the parent nucleus must be greater than that of the daughter nucleus by at least the mass m of the positron it creates. The mass of the parent atom must be greater than that of the daughter by at least $2mc^2 = 1.022$ MeV.

A positron slows down in matter and then annihilates with an atomic electron, giving rise to two photons, each having energy $mc^2 = 0.511$ MeV and traveling in opposite directions. The 0.511MeV annihilation photons are always present. In addition, because of the competing process of electron capture, characteristic X rays can be expected [34].

PAS is used to investigate changes in topaz samples before and after gamma irradiation. The most recent work of a positron lifetime investigation showed that the vacancy clustering signal is associated with a modest increase in positron lifetime and a large increase in the positron trapping rate [39].

The annihilation characteristics of trapped positrons reflect local properties of defects. It is assumed that in a perfect crystal delocalized positrons annihilate with a single lifetime, the so called bulk lifetime $\tau_b=1/\lambda_b$. In the presence of defects, positrons may get localized at them and annihilate with a second lifetime $\tau_d=1/\lambda_d$.

2.8.3. Basic principles of positron annihilation

Dirac (1930) predicted the existence of a positron in his relativistic theory. Then three years later, Anderson (1933) confirmed the existence of the positron

experimentally in his studies of cosmic radiation. Positron has the same mass and spin as electron but has opposite charge and magnetic moment.

The technique of (PAS) relies on the fact that a positron that comes in the immediate vicinity will cease to exist by annihilation. In the annihilation process of a positron and the electron gamma photons are set free that can be detected. If positrons are injected into a solid body their lifetime will strongly depend on whether they end up in a region with high electron density or in a void where electrons are scarce or absent. In the latter case the lifetime can be much longer because the probability to run into an electron is much lower.

By comparing the fraction of positrons that have a longer lifetime to those that annihilate quickly one can therefore gain insight in the voids or the defects of the structure.

Positrons emitted from a radioactive source, interact with matter in the same manner as electrons. A positron loses rapidly its kinetic energy through elastic and inelastic collisions with electrons as well as via phonon scattering and becomes thermalized. The time needed for thermalization is short ($\sim 10^{-11}$s) compared to the mean lifetime of positrons in metals (10-50) x 10^{-11}s. The thermalized positrons will then diffuse in the material until it finally annihilates with electrons [38]. In this process, the mass of the particle-antiparticle is converted into gamma radiation ($E=2mC^2$) in the form of two photons each of energy 511 KeV, which are emitted at 180° owing to conservation of energy and momentum, respectively. The positron annihilates with an electron according to the reaction:

$$e^+ + e^- \rightarrow 2\gamma \qquad (2.8)$$

The technique requires a source of positrons, where a radioactive isotope of sodium is often used. Therefore, the study of the characteristics of the annihilation photon will gives information about the state of the annihilation pair just before the conversion process. Figure (2-3) shows schematically the

positron annihilation, where the most commonly used positron isotope ^{22}Na is applied.

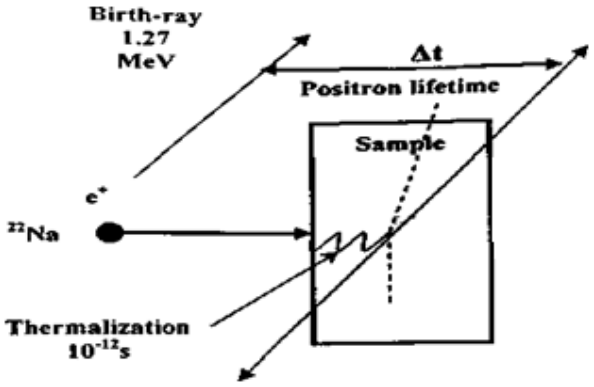

Figure (2.3); Scheme of positron annihilation

2.8.4. Positron Annihilation Process in Materials

The annihilation radiation gives detailed information on electron density and momentum distribution in the region scanned by positrons. The theoretical lifetime spectrum can be described by Eq.(2.8) (Jean and Scharder(1988) [(40)]:

$$\lambda (t) = (I/\tau)\, e^{-t/\tau} \quad \rightarrow \qquad (2.9)$$

Where, λ, is the decay rate, I is the positron intensity and τ is the mean lifetime. When positrons annihilate from a number of discrete states N the equation (2.9) changes into:

$$\lambda (t) = \sum N \ (Ii/\tau i)\, e^{-t/\tau_i} \quad \rightarrow \qquad (2.10)$$

Where τ_i is the positron mean lifetime in "I"th state and I_j is the corresponding intensity. These two values yield information on the local electron density and the probability of population of a specific trapping sites, respectively.

The positron lifetime varies only from 170 ps in aluminum to 420 ps in Cesium, where the free electron density decreases by a factor of 20 [(Springer and Radomsk 1998)] while plastic deformation of over-aged aluminum alloy at room

temperature increases the average positron lifetime from initial value of 190 ps to 203 ps [41]. It was noticed that, deformation or heating of the sample to elevated temperatures causes remarkable changes in the annihilation characteristics. In defects, where atoms are missing or their density is locally reduced, the repulsion between the positron and ion cores is decreased. Also, the redistribution of electrons causes negative electrostatic potential at this type of defect. Thus positrons see defects like vacancies, voids and dislocations as strongly attractive centers in the crystal. Theoretical calculations have been done to establish correlation between the positron lifetime "t" in defects and the vacancy size R of the clusters, which are assumed to be of spherical shape {(Hautojarvi et al. (1977)} [42].

On the other hand, the concentrations of defects can be deduced from the ratio of trapped and free positrons. This means that, the annihilation characteristics of trapped positrons reflect local properties of defects especially those, which are too small (10~9 m) to be detectable by any other methods.

The ^{22}Na isotope gives a relatively high positron yield of 90.4 % and has several other advantages. In addition to ^{22}Na, other isotopes like (^{64}Cu, ^{58}Co, etc.) can be used, but are less common. Also pair production resulted from bremsstrahlung radiation produced by high energy electrons consider another example for obtaining Positrons.

In the present work, the radioisotope employed is Na. The reasons for using ^{22}Na source are based on the following arguments:

1. The long lifetime of ^{22}Na radioisotope; (2.6 years).
2. It has a prompt γ-ray (1.28 MeV) accompanying the positron emission, which is distinguishable from the annihilation y-ray (0.512MeV). This prompt γ-ray is used as a signal for the birth of the positron whereas the annihilation γ-ray is signaling its death.
3. The probability of positron decay of ^{22}Na to the first exited state at 1.28 MeV is 90% of the total decay.

4. Its price is reasonable.
5. The accidental contamination of laboratory personnel is less harmful, since the biological half-life is only a few days.

Sources of weak activity are needed for positron lifetime even activities as low as ($7.4x10^5$) Bq [(20 μCi)] are sufficient. The sources are usually prepared by evaporating a solution of a ^{22}Na salt on a thin metal or polymer foil. The most common foil materials are Al, Ni, and mylar or kapton. In order to ensure the almost complete annihilation of positrons in the specimen volume, the foil source is placed between two identical samples (sandwich arrangement). A minimum sample thickness is required to ensure that the essential fraction of positrons annihilates in the sample pair (~2mm). The scintillators should be placed as close as possible to the sample–source sandwich. If cooling of the sample is required, the design of the cryostat should allow the separation between the scintillators to be as small as possible in order to avoid a considerable reduction in the counting rate. A small fraction of the positrons annihilates in the source. This amounts to 2 to 15 % depending on the foil thickness and the atomic number of the sample, which determines the backscattering and, thus, the multiple passing of positrons through the source. For the analysis of positron lifetime spectra, this fraction must be carefully determined and subtracted. The so-called source correction is an essential task of the lifetime spectra evaluation. To reduce the source contribution, the isotope solution can be directly deposited onto the sample surface. The disadvantage of this procedure is radioactive contamination of the samples, which can thus hardly be used for further investigations.

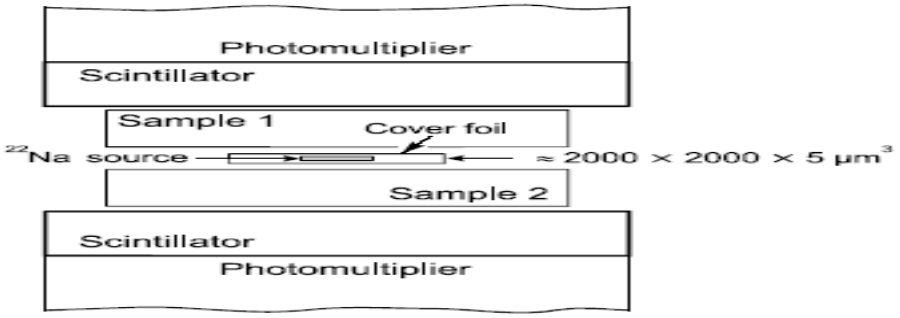

Figure (2.4); Scheme of the sample–source sandwich arrangement

The previous figure shows an arrangement for the registration of the birth and annihilation γ-rays as start and stop signals in the conventional positron lifetime experiment. The γ-quanta are detected as light flashes in the scintillators. These photons are converted into electrical pulses in fast photomultipliers. ^{22}NaCl is typically used as the source material.

2.8.5. Sample requirements

Two identical samples for the measurement are necessary. The samples will be measured in "Sandwich Geometry", i.e. they surround the ^{22}Na-source which is packed into thin (about 2 mm) Al foil.

The samples should be larger than the source, so that the minimum size is about 3 x 3 mm. In order to ensure that 99.9% of the positrons will stop and annihilate in the sample, a certain sample thickness is required. It is called "information depth". The surface quality is not very important, but the surface should not be damaged.

2.9. Radiation Protection

Radiation protection, sometimes known as radiological protection, is the science of protecting people and the environment from the harmful effects of ionizing radiation, which includes both particle radiation and high energy electromagnetic radiation.

Ionizing radiation is widely used in industry and medicine, but presents a significant health hazard. It causes microscopic damage to living tissue, resulting in skin burns and radiation sickness at high exposures and statistically elevated risks of cancer, tumors and genetic damage at low exposures.

2.9.1. Objective of Radiation Protection Program

The main objectives of the radiation protection program are

A- To prevent deterministic effects due to ionization radiation, and

B-To reduce the probability stochastic effects due to ionizing radiations

The objective of radiation protection program is implemented through applying elements of radiation protection program.

Among these elements the following: - [43]

1- Establishment of radiation protection infrastructure
2- Proper selection of personnel and training
3- Occupational Control
4- Public control
5- Emergency program
6- Quality Control of Radiation Protection Program

2.9.2. Radiation Protection infrastructure [44]

2.9.2.1. National Infrastructures

A national infrastructure is in place enabling the Government to discharge its responsibilities for radiation protection and safety. Essential parts of a national infrastructure are: legislation and regulations; a Regulatory Authority empowered to authorize and inspect regulated activities and to enforce the legislation and regulations; sufficient resources; and adequate numbers of trained personnel. The infrastructures must also provide ways and means of addressing societal concerns which extend beyond the legal responsibilities of the legal persons authorized to conduct practices involving sources of radiation. For example, national authorities ensure that appropriate arrangements are made for detecting any buildup of radioactive substances in the general environment, for disposing of radioactive wastes and for preparing for interventions, particularly during emergencies that could result in exposure of the general public. They also need to provide for the control of sources of radiation for which no other organization has responsibility, such as natural sources and radioactive residues from past practices.

National infrastructures must provide for adequate arrangements to be made by those responsible for the education and training of specialists in radiation protection and safety, as well as for the exchange of information among specialists.

A related responsibility is to set up appropriate means of informing the public, its representatives and the information media about the health and safety aspects of activities involving exposure to radiation and about regulatory processes. This provides information to facilitate the political process of setting national priorities and allocating resources for protection and safety and also helps to make the regulatory process more readily understandable.

National infrastructures must also provide facilities and services that are

essential for radiation protection and safety, but are beyond the capabilities required of the legal persons who are authorized to conduct practices. Such facilities and services include those needed for intervention, personal dosimetry and environmental monitoring, and for calibration and intercomparison of radiation measuring equipment.

Services could include the provision of central registries for occupational exposure records and the provision of information on equipment reliability. The provision of such services at the national level does not detract from the ultimate responsibility for radiation protection and safety borne by the legal persons authorized to conduct the practices.

Sources and practices involving the potential exposure of humans to ionizing radiation are normally controlled by a system of notification and authorization [(45)]. Regulatory systems for radiation protection are intended to ensure the Protection of people from harm arising from exposure to ionizing radiation is the first main. However, there are some human activities involving exposure to radiation that do not warrant regulatory control. Such circumstances arise when the resources that would need to be expended in regulating the activity would be excessive in relation to any benefit that might ensue in terms of reduced risk [(46)].

The IAEA issued Safety Series 115, "International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources", commonly known as the (BSS) in 1996; and it is consider as a main reference at this field. The BSS indicate that radioactive material from an authorized practice or source whose release to the environment has been authorized is exempted from any new requirements of notification, registration or licensing unless otherwise specified by the regulatory body. Since exemption and clearance are in essence generic authorizations, this provision of the BSS means that ‘exempted’ or ‘cleared’ material should be allowed to be used without any further restriction; this means that material that has been exempted

or cleared should not reenter the system of protection for practices, unless the regulatory body specifically requires that it do so.

Radioactive material categorized into two groups; Exclusion and regulatory control which divided to four groups; authorized discharge, authorized disposal, Exemption and clearance.

The BSS [44] establish the requirements for protection against the risks associated with radiation exposure. In spite of the low radiation levels, blue topaz is radioactive material and has to be regulated and controlled as such [47]. The BSS cover both practices and interventions and present the concepts of exclusion, exemption and clearance [48]. The BSS require national authorities to foster and maintain a safety culture. To this end all persons associated with radiation work have to be suitably trained and qualified so that they understand their responsibilities and perform their duties safely [49].

Persons who work in radiation protection field are categories to: qualified experts, radiation protection officers, workers, including personnel working directly with sources of radiation and those persons with a low potential for exposure, qualified operators, health professionals, employers, registrants and licensees, staff of regulatory authorities and emergency response personnel [50].

Some types of sources of ionizing radiation are not subject to regulatory control, either because they are not amenable to such control (e.g. cosmic rays) and are therefore excluded from the regulatory process, or because they present such a low risk that control by regulatory processes would be a waste of resources. In the latter case, two categories can be distinguished:

(a) Radiation sources which never enter the regulatory control regime, i.e. control is not imposed, and

(b) Radiation sources which are released from regulatory control; control is removed.

Sources in the first category are excluded from regulatory control by a process called exemption. Exempted sources typically include small sources of radiation such as tracers used in research, calibration sources and some

consumer products containing small sources or low levels of activity per unit mass. The corresponding levels of activity or activity concentration are called exemption levels. In the second category the release of sources from control is called clearance. Cleared sources include waste materials and materials for recycling from within the nuclear fuel cycle and wastes from other regulated facilities such as hospitals, research laboratories and industry where treated gemstones could be added to it. The amounts of material involved in clearances can be substantial and are generally greater than those involved in exemptions of man-made sources. The corresponding levels of activity or activity concentration are called clearance levels [(45)]. These concepts and the relations between them are briefly described.

2.9.3. Recommendations of the International Commission for Radiological Protection (ICRP)

ICRP recommendations were developed from Dose limitation system to protection in practice and protection at intervention to 2007 recommendations.

2.9.3.1. System of dose limitation

Following ICRP-26 (1977), the system of dose limitation was established where the elements of dose limitation system are:

A- Justification of practice

B- Optimization for protection and

C- Annual dose limits

IAEA issued SS-9 (1982) and in United Kingdom Ionizing radiation regulation (IRR-8)5 was issued.

2.9.3.2. System of Protection in practice

As a result of Major international Accidents such as Chernobyl accident, ICRP modified its recommendations to ICRP-60.

Following ICRP-60 (1990), the system of protection at practice was established with three elements, these are

A- Justification of practice

B- Optimization for protection and

B- Dose limits

IAEA with international organizations issued SS-115(1996), and in United Kingdom Ionizing Radiation regulation (IRR99) was issued.

2.9.3.3. System of protection at Intervention

Following ICRP-60(1990), the elements of protection at Intervention are

A- Justification for intervention, and

B- Optimization or intervention

No dose limits or intervention.

2.9.3.4. Latest 2007 ICRP recommendations

Following latest ICRP recommendations adopted in 2007, and issued as ICRP-103 in 2008 a unified system of protection was developed, from exempted practice, occupational, emergency.

IAEA with international organizations are Revising BSS (SS-115) and the work is still in progress.

In order to proper implement radiation protection program radiation measuring devices are needed.

2.9.3.4.1. 2007 ICRP Recommendations Dose Limits

1. 2007 ICRP recommendation dose Limits apply only in planned situations. And it defined three types of exposure; these are occupational, public exposures well as medical exposures. In the present study two types of exposures are considered, occupational for workers handling neutron irradiated Gemstones and public who use it such as house wives.
2. For Occupational exposure in planned situations the limit should be expressed as an effective dose of 20 mSv in a year, averaged over defined 5 years periods (100 mSv in 5 years), with provision that the effective dose should not exceed 50 mSv in any single year.
3. For public exposure in planned situations the limit should be expressed as an effective dose of 1 mSv in a year.

4. The limit on effective dose applies to the sum of external exposures and internal exposures due to intakes of radionuclides.
5. There are recommended dose limits in planned exposure situations must be considered.
    a) Dose limits to lens of the eye (150 and 15) mSv.
    b) Dose limits to skin (500 and 50) mSv.
    c) Dos limits to hands and feet (500 and nothing) mSv.

    All these limits are for occupational and public respectively.

2.9.4 Radiation Quantities and Units (ICRP 103) [51]

The following quantities and units are in use for radiation protection purposes

* Radioactivity.
* Absorbed dose.
* Equivalent dose.
* Radiation weighting factor.
* Effective Dose.
* Tissue weighting actor.
* Collective dose.

2.9.5. Unit of Radioactivity

The main physical quantities used in the standards are the rate of nuclear transformation of radionuclides (the activity) and the energy absorbed by a unit mass of a substance from the radiation to which it is exposed (the absorbed dose). The unit of activity is the reciprocal second, representing the number of nuclear transformations (or disintegrations) per second, which is termed the Becquerel (Bq), defined as one disintegration per second: 1 Bq = 1 s^{-1}. The unit of absorbed dose is the joule per kilogram, termed the gray (Gy). The absorbed dose is the basic physical dosimetric quantity of the Standards [44]. The traditional unit of activity is the curie (Ci), which was originally the activity

ascribed to 1 g of ^{226}Ra. The curie is now defined as 1 Ci = 3.7×10^{10} Bq, exactly.

2.9.6. Exclusion, Exemption and Clearance

2.9.6.1 Introduction

Recent publications (2003) by IAEA were issued related to the concepts of exclusion, exemption and clearance. Furthermore, exclusion, exemption and clearance principals were included in latest 2007 ICRP recommendations. In the present study, application of the system of exclusion, exemption and clearance to neutron irradiated gemstones. Gemstones contain impurities which are made radioactive during neutron exposure. Hence radioactivity is introduced.

The dose delivered to these gemstones is an important consideration in studying induced activity. It is necessary to allow for radioactivity to decay before the irradiated gemstones can be handled.

2.9.6.1.1. For occupational worker

Exposure of the individuals that retrieve and work with irradiated gemstones, immediately after an irradiation, must be maintained within the occupational exposure limits. The level of their exposure to radiation will depend on several factors, including:

(a) The type of treatment.

(b) The neutron (radiation) fluence delivered to the gemstones, and

(c) The nuclides and activity levels created by the neutrons irradiation.

If the radiation protection principles of time, distance, and shielding are followed, it is anticipated that the exposure of the individuals handling gemstones immediately after irradiation can be maintained within occupational exposure limits [(9)].

Optimization for protection to radiation workers is achieved through distance, time and shield.

2.9.6.1.2. For members of the public

Gemstones must be cleared before distribution. This means that its exposure to members of the public must be less than cleared level [10 micro Siveret (μSv) per Year]

2.9.6.2. Definitions:-

2.9.6.2.1. Exclusion

The BSS state that "Any exposure whose magnitude or likelihood is essentially unamenable to control through the requirements of the standards is deemed to be excluded from the standards". Examples of excluded exposure given in the BSS are: "exposure from ^{40}K in the body, from cosmic radiation at the surface of the earth and from unmodified concentrations of radionuclides in most raw materials".

2.9.6.2.2. Exemption

The BSS use the concept of exemption only within the context of practices and sources within these practices. Exemption determines a priori which practices and sources within practices may be freed from the requirements for practices on the basis of their meeting certain criteria. Exemption may be considered a generic authorization granted by the regulatory body which releases the practice or source according to justification principle. There are levels for exemption dependent on the activity concentrations and total quantities of radionuclides.

2.9.6.2.3. Clearance

Clearance defined as the removal of radioactive material or objects from any further regulatory control by a regulatory body. Clearance of bulk amounts of materials with activity concentrations lower than the guidance exemption levels specified by specialist in references are require further consideration by the regulatory body [48]. Regulations containing specifications for exemption or clearance levels relevant to the application of radionuclides in hospitals,

research institutes and industry exist in many industrialized countries but the radiological basis for the values in national regulations is often not well defined and the values differ from one country to another [45].

From the previous definitions, Exclusions principal can not be applied to workers and public handling Gemstones.

If the radioactivity concentration due to irradiations of gemstones to neutrons exceeds the exemptions level, the work place should be licensed, while if the radioactivity concentration is less than the exemption level, then the workplace is exempted. Furthermore, the neutron irradiated gemstones can only be handled to member of the public when the radioactivity concentration is less than the clearance level.

2.9.7. Decay storage:

Storage of radioactive material prior to clearance in order to allow decay of short-lived radionuclides must be done [52]. Values of activity concentration radionuclides of natural origin are shown at the table (2.2).

Table (2.2); values of activity concentration for radionuclides of natural origin

Radionuclide	Activity concentration (Bq/g)
^{40}K	10
All other radionuclides of natural origin	1

2.9.8. Radionuclides of Artificial Origin

The values of activity concentration for bulk amounts of material containing radionuclides of artificial origin, derived using the exemption concept are given in the table (2) at the safety guide [48] and here some radionuclides related to this work will be showed at the table (2.3).

Table (2.3); Activity concentration values for bulk amounts of some radionuclides of artificial origin [46]

No.	Radionuclide	Activity concentration (Bq/g)
1	Na-24	1
2	Sc-46	0.1
3	Mn-54	0.1
4	Fe-59	1
5	Co-60	0.1
6	Zn-65	0.1
7	Zr-95	1
8	Ag-110m	0.1
9	Sb-124	1
10	Cs-134	0.1
11	Eu-152	0.1

In the Safety Guide, the concepts of exemption and clearance have been applied to bulk amounts of material containing radionuclides of artificial origin. These concepts relate specifically to practices that are considered by the regulatory body to be justified. If the activity concentrations of radionuclides in materials are below the values of activity concentration given in table 2 at the safety guide [48], the handling and use of the material may be considered exempt from the regulatory requirements for practices.

2.9.8.1. Application of the Radionuclides Values of Natural Origin

It is usually unnecessary to regulate radioactive material in activity concentrations below the values given in the reference table. However, there are some situations which exposures from materials due to radionuclides with activity concentrations below those given in the reference table would necessitate consideration by the regulatory body for some types of regulatory control.

If the activity concentration of the radionuclide exceeds the value of activity concentration given in the reference table, the regulatory body should decide on the extent to which the regulatory requirements set out in the BSS should be applied.

2.9.8.2. Trade

If the values of activity concentration provided in the safety guide no need for any further action for materials containing radionuclides at activity concentrations below these values. In particular, national and international trade in commodities containing radionuclides with activity concentrations below the values of activity concentration provided in the two tables mentioned before should not be subject to regulatory control for the purposes of radiation protection. One of the requirements in the new standards is that the disposal, recycling and reuse of material containing radioactive substances is subject to prior authorization by national competent authorities. It is stated, however, that the authorities may specify clearance levels below which such materials are no longer subject to the requirements of the Standards [53]. Treated gemstones as previous radioactive materials must be clearance with levels below it these treated gemstones are no longer subject to any requirements of the standards and become free for use. Three sets of clearance levels were developed [54]:

1. For Reuse (or Demolition).
2. For Demolition only.
3. For Building Rubble.

Until this date there is no data about clearance levels of treated gemstones, but it assumed that it will be treated in the same manner because the clearance levels derived are for unrestricted release [54].

2.9.8.3. Application of Clearance Principles

2.9.8.3.1. The need for derived quantities

The guiding radiological criteria for exemption and clearance are expressed in terms of dose and cannot be used directly for establishing exemption or clearance levels. Hence it is necessary to convert them into practical quantities. For solid materials useful quantities are mass activity concentration (Bq/g), surface contamination (Bq/cm^2) and total activity per unit time (Bq/s).

There is a reference table contains derived values for clearance from regulatory control of solid materials. This table specifies radionuclide, ranges of activity concentration (Bq/g) and representative single values of activity concentration (Bq/g) [45]. The clearance levels are then 0.3, 3, 30, 300 and 3000 Bq/g for the five classes.

In the absence of other guidance, the clearance level values for surface contamination (Bq/cm^2) may be taken to be the same in unit terms as for activity concentration (Bq/g) [45].

Table (2.4); Derived values for clearance from regulatory control of solid materials.

radionuclide	ranges of activity concentration (Bq/g)	representative single values of activity concentration (Bq/g)	Class number
Na-24	0.1→<1.0	0.3	1
Mn-54	0.1→<1.0	0.3	1
Co-60	0.1→<1.0	0.3	1
Zn-65	0.1→<1.0	0.3	1
Ag-110m	0.1→<1.0	0.3	1
Sb-124	0.1→<1.0	0.3	1
Cs-134	0.1→<1.0	0.3	1
Fe-59	≥1 → < 10	3	2

Table (2.5); Isotopes activity for Bulk (with different weight)

radionuclide	ranges of activity concentration (Bq/g)	representative single values of activity concentration (Bq/g)	Class number
Na-24	0.1→<1.0	0.3	1
Mn-54	0.1→<1.0	0.3	1
Co-60	0.1→<1.0	0.3	1
Zn-65	0.1→<1.0	0.3	1
Ag-110m	0.1→<1.0	0.3	1
Sb-124	0.1→<1.0	0.3	1
Cs-134	0.1→<1.0	0.3	1
Fe-59	≥1 → < 10	3	2

Treated gemstones should be deal as Building materials which exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at the most. This is the excess gamma dose to that received outdoors [(55)].

2.9.9. Transportation

2.9.9.1. Introduction

Radiation and radioactive substances have many beneficial applications. There is high necessity for transport it from its original place to other places. Gemstones are good example for the necessity of transportation. The transportation of radioactive materials is controlled by finite regulations. The radiation risks to workers, the public and to the environment that may arise from these applications have to be assessed and, if necessary, controlled. Activities such as the medical uses of radiation, the operation of nuclear installations, the production, transport and use of radioactive material, and the management of radioactive waste must therefore be subject to standards of safety [(56)]. Transportations for radioactive materials inside the work place must be also controlled by local regulations.

The activity per gram of the gemstones per unit fluence varied widely for any particular country of origin, sometimes by several orders of magnitude. This variation was due to the differences in the amount of specific precursor nuclides in the individual topaz. For this reason alone, recommendations can not be made about limiting or encouraging using topaz from a specific country of origin to minimize activity levels. Although the amount of precursor nuclide varied greatly, the distribution of activity in an individual topaz was uniform [(9)].

2.9.9.2. Working with radioactive materials:

Radioactive Material(RM) means any material containing radio nuclides where both the activity concentration and the total activity in the consignment exceed the values specified in paragraph "401-406,ST-1" [(57)]. Any use of unsealed radioactive materials should be undertaken on the assumption that contamination may occur. Forethought and planning before operations commence will greatly facilitate clean up operations after any contamination occurs [(58)]. Special form radioactive Material (SFRM) means either an

indispensable solid radioactive material or a sealed capsule containing radioactive material [57].

Safe transport of radioactive material regulations issued by IAEA since 1961, provide standards for insuring a high level of safety of people, transport workers, property and environment against radiation, contamination and criticality hazards as well as thermal effects associated with the transport of the radioactive wastes and material [59].

The use of radioactive material nowadays becomes very important in most fields; such as medicine, industry, agriculture, research, consumer products and electrical power generation .Tens of millions of packages containing radioactive material are consigned for transport each year throughout the world [60]. The quantity of radioactive material during transportation changes from very small to high big.

2.9.9.3. Description of regulations

The Regulations for safe transport of radioactive material establish standards of safety which provide an acceptable level of control of the radiation with the transport of radioactive material [61]. These Regulations depends on the principles of the "Radiation Protection and the Safety of Radiation Sources" [61] and the "International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources" [44] as international regulations. Each country establishes specific regulations according to its laws but it must be agree with international regulations. Gamma-irradiated gemstones are not byproduct materials unless the gamma energy exceeds the threshold energy for activation [62].

The transport of radioactive material by any person, organization or government must comply with the radiation safety legislation of the State, territory or Commonwealth jurisdiction through which the radioactive material is transported [57]. From 1957 up now IAEA develop its regulations for safe transport of radioactive materials. The objective of these Regulations is to

establish requirements that must be satisfied to ensure safety and to protect persons, property and the environment from the effects of radiation in the transport of radioactive material. This protection is achieved by requiring:

(a) Containment of the radioactive contents;

(b) Control of external radiation levels;

(c) Prevention of criticality; and

(d) Prevention of damage caused by heat [56].

The regulations could be achieved by applying quality assurance program. It is applied to:

a) The transport of radioactive material by all modes on land, water or in the air,

b) Any transport which is incidental to the use of the radioactive material.

Transport comprises all operations and conditions associated with, and involved in, the movement of radioactive material; these include:

The design, manufacture, maintenance and repair of packaging, and the preparation, consigning, loading, carriage including in-transit storage, unloading and receipt at the final destination of loads of radioactive material and packages.

Regulations are characterized in terms of three general severity levels [56]:

(a) Routine conditions of transport (incident free);

(b) Normal conditions of transport (minor mishaps);

(c) Accident conditions of transport.

2.9.9.4. Categories for Packages and Overpacks:

Overpack (OP) means an enclosure such as a box or bag. With regard to the shielding provided, packages are classified in three categories, I-White, II-Yellow and III-Yellow. Each is associated with specified maximum radiation levels at the external surface of the package [56].

Table (2.6); Categories of the packages and overpacks [56].

Transport Index (TI)	Maximum radiation level at any point on external surface	Category
0	≤ (0.005 mSv/h)	I-White
0 → 1	>(0.005 mSv/h) ≤(0.5 m Sv/h)	II-Yellow
1→ 10	> (0.5 m Sv/h) ≤ (2 m Sv/h)	III-Yellow
>10	> (2m Sv/h) ≤ (10m Sv/h)	III-Yellow

TI: The maximum radiation levels at a distance of 1 meter from the external surface. It is determined by multiplying the radiation level at one meter by 100.

Most of the pure alpha and beta-emitters could be transported as white packages, it would be economical to transport gamma emitters as yellow packages. Otherwise considerable amounts of shielding would be required to bring the radiation levels to those corresponding to white packages .All fissile materials are packed and shipped in such a manner that criticality cannot be reached under any foreseeable circumstances of transport. Transport container named Freight Container (FC)Meant an article of transport equipment designed to facilitate the carriage of goods either packaged or unpackaged. A small FC is that which has any overall outer dimension less than 1.5 m, or an internal volume of not more than 3 m^3.

2.9.9.5. Occupational exposure arising from transport activities

The occupational exposure arising from transport activities, where it is assessed that the effective dose:

* Is most unlikely exceed 1mSv in a year, neither special work patterns nor detailed monitoring nor dose assessment programmer nor individual record keeping shall be required;

* Is likely to be between 1 and 6 m Sv in a year, a dose assessment programmer via work place monitoring or individual monitoring shall be conducted;

* Is likely to exceed 6 m Sv in a year, individual monitoring shall be conducted.

When individual monitoring or work place monitoring is conducted appropriate records shall be kept.

2.9.9.6. Controls for Contamination and Leaking Package

Non–fixed radioactive contamination (NFRC) means contamination that can be removed from a surface by wiping with a dry smear. The non- fixed contamination on the external surface of packages shall be kept as low as practicable and under conditions likely to be encountered in routine transport, shall not exceed the following levels is 4 (Bq/cm^2) for Beta and gamma emitters and low toxicity alpha emitters and 0.4 (Bq/cm^2) for all other alpha emitters. The same limits apply to the external and internal surfaces of overpackes, freight containers, tanks and intermediate bulk containers.

Radioactive contamination may result from the use of unsealed radioactive material in industrial, scientific or medical laboratories, and includes contamination of designated radioisotope areas and of work areas which are not designated as radioisotope areas but which may inadvertently become contaminated with unsealed radioactive material [58].

The surface contamination limits are based on the committed effective dose limit of 20(mSv) per year recommended for workers by the National Health and Medical Research Council "NHMRC" [63].

2.9.9.7. Contamination [4]

* Most stones are cut and polished prior to irradiation.
* NO stones are cut or polished by tools leave traces.
* In addition, any items that are to be electroplated must be rigorously cleaned to ensure proper adhesion of electroplated metals.
* Accordingly, there should be no contamination.

This should be checked by manufacturer, prior to shipping.

2.9.9.7.1. Types of surfaces [58]

Limits are presented for surfaces in work areas which are designated for the use of radioisotopes; surfaces include protective clothing worn by workers. Modifying factors are presented for:

• The skin of workers;

• Personal clothing;

• Work areas not designated as ones in which unsealed radioactive material may be used; and

• The interiors of glove boxes and fume hoods.

2.9.9.8. Results and Discussions:

Gem stones are those stones which have beauty that can be used based on its color, transparency and brilliance. Treated gem stones have extremely valuable. Therefore, it is considered as gemstone after irradiation by neutrons or gamma rays [28].

Treated Gem stones have extremely valuable in jewelers, research and industry. Nowadays, large quantities are produced by several specialized centers scattered allover the world. The number of consignments of radioactive materials being transport within countries and between different countries is likely to increase with the development of nuclear power and the use of other nuclear techniques. Accidents can happen in all modes of transport, and it is likely that a significant number of accidents will involve packages of radioactive materials.

2.9.9.9. Condition of transport of previously irradiated stones to abroad [28]

The transport of radioactive material must be subject to a radiation protection program, which must consist of systematic arrangements aimed at providing adequate consideration of radiation protection measures. The nature and extent of the measures to be employed in the program must be related to the magnitude and likelihood of radiation exposure.

The main residual radioactivity is due to Sc-46, Ta-182, Mn-54 and other isotopes which are equal to about 56 (Bq/g) which represents 75.67% of the IATA permissible level. Thus it could be transport without any problems [28].

2.9.9.10. Transport Regulation

Competent authority means any national or international regulatory body or authority designated or otherwise recognized as such for any purpose in connection with these regulations.

2.9.9.10.1. Egyptian National Regulations

The competent authority is responsible for providing the national regulations, which implement the IAEA Regulations and for up-dating them when necessary. The national regulations must fully and accurately reflect the requirements of the IAEA Regulations. This is important to ensure harmony with other countries Regulations. Also, the national regulations must suitably extend the IAEA`s Regulations to cover all national needs and all practical operation requirements relating to modes involved. These regulations must be followed at all nuclear facilities inside the country.

2.9.9.10.2. International Regulations

In general, the organization of a national input to IAEA reviews or other regulatory revisions involves administrative action possibly supplemented by technical contributions.

Technical contributions are required in regulatory reviews. So it is necessary to arrange for national representation at international regulatory meetings. International Organizations interested by the field of transportation are UN Committee of Experts, International Civil Aviation Organization (ICAO), International Maritime Organization (IMO), Inter-Agency co-ordination activities, European Commission, ADR/RID, IATA, IFALPA and ISO [(64)].

The overall objective of our study was to quantify the concentrations of activity possible in irradiated gems, to identify the specific nuclides responsible for the radiations, and to measure or calculate the dose to people attributable to the induced activity. Such information was especially incomplete for pure beta emitters, because they present a difficult analytical challenge in the presence of nuclides that emit both beta particles and gamma rays. In this study, the types of pure beta emitting nuclides that could be expected were identified and their activity concentrations were measured in activated topaz. Such analyses are

essential in evaluating the health risk associated with distributing and wearing such gems.

Another factor investigated was the distribution of the activity in the gem. If the induced activity was not uniformly distributed, then certain beta emitting nuclides could go undetected yet still result in concentrations exceeding the exempt concentration limits.

Irradiated topaz gems from several countries with neutrons in a high-flux nuclear reactor to maximize the induction of radioactivity allowing measurements to be completed in a reasonable time are widely used nowadays. Worst case scenarios were completed on which to formulate upper estimates of health risks and to make correlations between the country of origin, type of emissions, and the magnitude of induced activity. Such correlations were considered to evaluate if one could safeguard occupational and public health on the basis of country of origin alone.

To determine if we could deduce activity concentrations from the color of topaz gems or the type of treatment, we compared the most commonly used coloration regimens. The identity and activity concentrations of radionuclides found with each type of treatment were established [9].

2.9.9.11. Transport Index (TI)

Transport index for a package means, the number expressing the maximum radioactive dose rate at distance of 1 meter from the external surface of the package. Or; the maximum radiation level in micro sievert per hour (μSv/h) at 1 m from the external surface of the package, divided by 10.

Example: 1μSv/h = 0. 1 mrem/h or Ti = 0. 1

2.9.9.11.1. Placement of Labels

The information and warning labels are displayed on two opposite sides of the outer surface of the container, not the side on which the container is to rest

or to be stacked during transport (do not place labels on top or bottom surface of container).

2.9.9.11.2. The United Nations Number

The United Nations Number (UNN) for the consignment shall also display on the placards (lower half) which shall be affixed adjacent to the main placard in all four sides of the freight container.

2.9.9.12. Low specific Activity material (LSAM)

Low specific Activity material (LSAM) by its nature has a limited specific activity of radioactive material. Surface Contaminated Object (SCO) means a solid object which is not it radioactive but which has radioactive material distributed on its surface. (SCO) is classified in one of two groups:

2.9.9.12.1. SCO-I; A solid object on which:

2.9.9.12.1.1. The non-fixed contamination on the accessible surface averaged over 300 cm^2 does not exceed 4 Bq/cm^2 (0.1nCi/ cm^2) for beta and gamma emitters and low toxicity alpha emitters, or 0.4 Bq/cm^2 (0.01nCi/ cm^2) for all other alpha emitters; and

2.9.9.12.1.2. The fixed contamination on the accessible surface averaged over 300 cm^2 does not exceed than 40 kBq/cm^2 (1μCi/ cm^2) for beta and gamma emitters and low toxicity alpha emitters, or 4 kBq/cm^2 (0. 1μCi/ cm^2) for all other alpha emitters [(65)].

2.9.9.12.2. SCO-II is a solid object on which either the fixed or non-fixed contamination on the surface exceeds the applicable limits specified for SCO-I [(65)]. The UN number for the bulk in this study is UN 2910 as illustrated at fig.

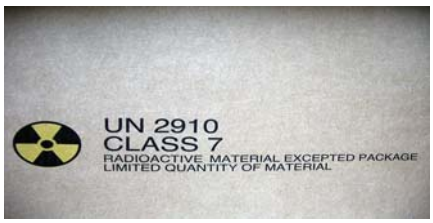

Figure (2.5); Information Label Hazard Labels [(65)]

A label with the proper TDG shipping name and UN number must be put. Shipments of dangerous goods are labeled in accordance with the Transportation of Dangerous Goods (TDG) Act. Labels allow the shippers, receivers and emergency response personnel to know the type of dangerous goods being handled as well as the relative risk. All packages must be categorized by radiation level and display radiation warning labels as follows:

2.9.9.13. Radiation warning labels

2.9.9.13.1. Category I

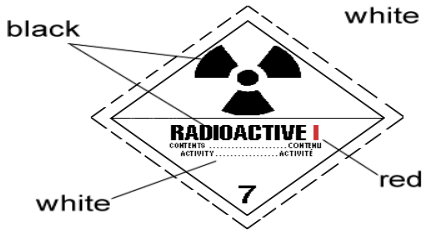

Figure (2.6); Category I label

The radiation level does not exceed than5 µSv/h at any location on the external surface of the package.

2.9.9.13.2. Category II

Figure (2.7); Category II label

The radiation level does not exceed than (500) µSv/h at any location on the external surface of the package and the transport index does not exceed 1.0.

2.9.9.13.3. Category III

Figure (2.8); Category III label

The radiation level does not exceed than (2) mSv/h at any location on the external surface of the package and the transport index does not exceed 10.

2.9.9.14. Dangerous goods classification:

There are nine classes. Material used at this study as radioactive material belongs to UN Class no "7". Dangerous Goods here which are treated gemstones have the previous symbols, and may divide to three categories.

Samples at this work considered as exemptions before transport and must to be clearance after finishing all processes and before distribution among users.

2.9.9.15. Storage of Radioactive Materials

Radioactive materials except those in category I- WHITE packages, shall be kept separated from living accommodations, from regularly occupied working spaces that may be continually occupied by passengers or the pubic.

CHAPTER (3)

Apparatus and Experimental Conditions

3.1 Topaz as a material under investigation

Topaz was selected because of its high desire in the gem trade market and for its experience (June1990) in the nuclear reactors [66].

All possible analyses were done during this study to ensure the optimum conditions to produce valuable treated gemstones with good benefit and to understand the coloration processes and the structure of the color centers.

Equipments used at this study could be separate into four parts according to its benefit in the study in the following:

(1) Equipments used for elemental analysis of natural topaz as (ICP-MS). It is used to investigate isotopes in the raw samples for the best choice of the samples which does not contain isotopes of long lived times.

(2) Equipments used as irradiators for topaz irradiation. Several types of irradiation can be used to alter the color in topaz as gamma rays from gamma facility and neutrons from reactor. Irradiation processes were carried out in the Egyptian second research reactor and the gamma facility at (AEA).

(3) Equipments used to study irradiated topaz. Neutron activation analysis is used to investigate the isotopes kind and its concentrations in the irradiated samples for the residual radioactivity calculation. Positron annihilation spectroscopy was used to study the induced defects by irradiation in topaz. Raman spectroscopy was used to study raw samples and irradiated stones to assist in the interpretation process of color at topaz. Spectrophotometer also was used to elucidate the changes occurred at the structure of treated gemstones through photoluminescence measurements.

(4) Radiation protection measuring devices which are divided into:

a) Equipment used for Monitoring of workplace.

b) Equipment used for storage of irradiated topaz.

c) Equipment used for transport of irradiated topaz.

Trace elements cause residual radioactivity in topaz after irradiation. It is not allowed to distribute topaz until it decays and reaches a safe level of use for the public. Radiation protection measuring devices are used to measure activity until it reaches the safety level for transportation of the samples. Also Storage of irradiated samples and isotopes activity for bulk were studied periodically. Also it used to achieve and clear radiation safety, health and safety, regulatory compliance including exempt distribution, exclusion, exemption and clearance. It is used before transportation to review (Transport Index "TI", United Nation Numbers, Dangerous goods classification).

In the following an overview of the apparatuses which were used in the measurements in this work:

3.2. Equipment:

3.2.1. Inductively Coupled Plasma Mass Spectrometer (ICP-MS)

High resolution inductively coupled plasma mass spectrometer (JMS-PLASMAX2) which is installed at the central laboratory for elemental and isotopic analysis, nuclear research center, atomic energy authority; Egypt is used in the analysis of the samples before irradiation.

3.2.1.1. The essential components of the ICP-MS are [28]:

1) An inlet-system for introducing the samples.

2) An ion source for producing the ion beam.

3) An analyzer by means of which the ion beam can be resolved into its various mass components.

4) A detecting system by means of which the resolved ion beams can be detected.

Figure (3.1); ICP-MS

3.2.1.2. General Features of the ICPMS [28]

The JMS-PLASMAX2 high-resolution ICP-MS, utilizing the latest model of the reversed double-focusing mass spectrometer equipped with quadruple focusing system and it has a resolution of 12000 [28]. A 40MHz high-frequency power supply and an automatic ignition system are used in the ICP torch. Ions introduced into the mass analyzer form the plasma through the plasma interface can easily be adjusted to form a high-intensity ion beam. The masses of the ion beam entering the double-focusing mass spectrometer are separated by the magnetic field and the electric field, and are detected by the post-accelerator analogue detector as well as acquisition and processing of the detected data are executed by the workstation system.

The plasma interface and the mass analyzer are separately evacuated by connecting them with a turbo-molecular pump (for high vacuum) and a rotary pump (for low vacuum), respectively. An air valve isolates the plasma interface and the mass analyzer. The air valve is opened and closed using argon gas.

3.3. Irradiation Facilities

Several types of irradiation can be used to alter the color in topaz: X-rays, gamma rays, neutrons, and high-energy charged particles such as electrons, protons.

Gamma and neutron irradiation can be conducted on quite large pieces of topaz; which is not true for high-energy-electron irradiation because of three factors; limited penetration depth, heat generation, and the build-up of electrical charge. Moreover, heating, too, cannot be performed on large topaz specimens without loss.

3.3.1. Gamma-Ray Facility

Gamma -1 unit of the national center of radiation research and technology was used. Cobalt-60 radioactive source was used as gamma source. The total activity at the time of the measurements was 3000 Ci (111TBq). The resultant gamma ray photons are with energies 1.17 MeV and 1.33 MeV, with average value of 1.25 MeV. Gamma rays are produced within a gamma cell. The rays are very penetrating and can produce uniform coloration if the material is uniform.

Figure (3.2); Gamma irradiation facility

3.3.2. Neutron Irradiation Facility

The Facility used for neutron irradiation is Egyptian second research reactor, ETRR-2. It is a multi purpose reactor, (MPR), a 22 MW power, open pool type, with maximum thermal neutron flux of 2.7×10^{14} $n.cm^{-2}.s^{-1}$. ETRR-2

is light water cooled and moderated, with Beryllium reflectors. Fuel elements are MTR type and 19.75 % enrichment. It aims to the production of radioisotopes for industrial and medical applications, research on neutron physics and personal training. Recently, facilities for gemstones irradiation have been introduced and fully utilized. The facility irradiates topaz stones to enhance its value for the users from industry. Specific irradiation container has been designed to ensure proper cooling of the stones during irradiations. Several irradiation tests have been conducted to define the optimum parameters of the facility. The neutrons in a nuclear reactor can be of varying energy and are also accompanied by gamma rays and other rays and particles. By placing the material to be irradiated into a cadmium-lined iron container, the thermal neutrons that do essentially all of the activating are absorbed by the metals, which then also generate additional gamma rays (Bastos, 1984)([67].

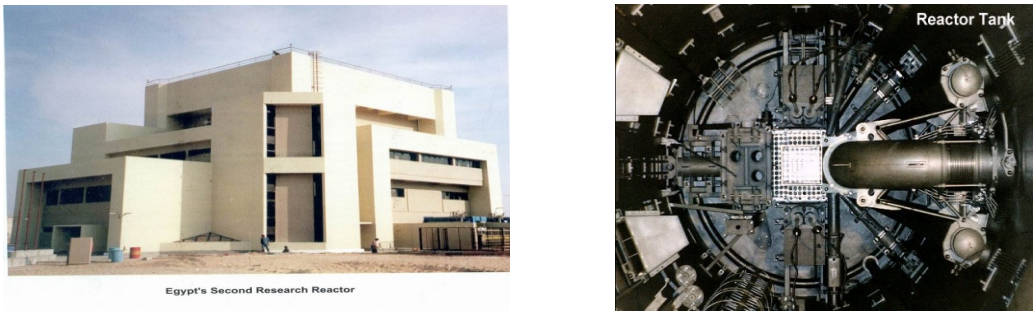

Figure (3.3); Reactor building and the reactor tank

3.4. Analysis after irradiation stage

3.4.1. Neutron Activation Analysis (NAA) Facilities

In the last years, neutron activation analysis has been applied for the analysis of treated gemstones. (NAA) is a very accurate and precise technique [68]. Short and long irradiations were implemented depending on the half-life of the induced radionuclides. The precision and accuracy of the method were tested by analyzing the certified reference materials. It is not easily affected by interferences if samples are properly prepared and in some cases permits the determination of elements hardly measured by other techniques [69]. An identical irradiation of samples, blanks and standards, must be assured. NAA is a technique in which gamma ray emissions are detected. Gamma ray emissions are usually distinctive enough that elements may be determined without chemical separations or special sample preparation. A sample is exposed to neutrons, resulting in activation of many of the constituent elements. Specific radiations emitted by the activation products are detected to determine the amount of the elements present in the sample.

3.4.1.1. Sample preparation

The samples are prepared for irradiation by the following procedure:

1- Weighing the sample.

2- Cleaning the sample using ultrasonic cleaner in trichloroethane followed by acetone (or ethyl alcohol) for about 10 minutes.

3- Etching the sample.

4- Water cleaning by ultra-pure water.

5- Drying the sample using nitrogen gas.

6- Cleaning the sample using ethyl alcohol.

7- Drying using nitrogen gas.

8- Weighing again.

After cleaning the samples they have to be prepared for irradiation as in the following steps:

1- Cover the sample with aluminium foil.

2- Put the sample in the sealed aluminium container.

3- Put the sealed aluminium container in the irradiation position.

4- Calculate the needed flux according to the needed dose.

5- Introduce this value to the shift supervisor.

6- Start the irradiation.

Samples of relatively uniform volume and mass are sealed inside polyethylene vials. Standards are similarly prepared. An irradiation time is selected, depending upon elements to be detected. Likewise, decay time and counting time are selected for the particular analysis desired. Standards and samples are counted on high purity germanium gamma ray spectrometers.

3.4.1.2. NAA Instrumentation

The gamma-ray spectroscopy system includes a computer, associated electronics, a lead shield that contains a detector, and a liquid-nitrogen storage tank. The heart of the system is the detector, which is made from a very-high-purity germanium (HPGe) crystal that is mounted inside an evacuated magnesium container (Debertin and Helmer, 1988; Knoll, 1989) [(70)]. The germanium crystal is cooled to nearly 77^0K (-196^0C) by the liquid nitrogen. To minimize the background radiation reaching the detector, the crystal is completely surrounded by a lead shield at least 10 cm thick. The shield is lined with cadmium and copper to prevent unwanted lower-energy gamma rays produced in the lead from reaching the detector.

Gamma rays from the irradiated gemstones are emitted randomly in all directions. Thus, when the stone is placed in position above the detector most of the gamma rays travel into the lead shield; only a small fraction enters the detector directly, and only some of these are completely absorbed and used in the analysis. However, when gamma rays are absorbed into the germanium crystal, they generate minute electrical pulses. These pulses are then amplified,

digitized, sorted by energy level, added, converted into spectrum peaks, and ultimately sent to the computer screen for visual analysis (NCRP, 1985) [71], [66].

Figure (3.4); HPGe system

3.4.1.3. Short Irradiation

For short irradiation, the sample was subjected to the following procedure: sample was accurately weighed into a polyethylene vials. After that, the polyethylene vials were marked, closed automatically then tested at liquid nitrogen and send to irradiation position for seconds or minutes using rabbit system may be for one time or many times continuously with fully control software system.

There are two computer pneumatic irradiation transfer systems. One position at the reflector area with thermal flux of $9x10^{13}$ n/cm^2/sec, and the other position at the thermal column with thermal flux of $6.8x10^{9}$ n/cm^2/sec. A supervision and control system with data acquisition and recording modules are available for the facility operation (samples ID, sending and receiving samples, time of flight accounting, auto-measure sample activity, and historical data.).

The rabbit travels from the laboratory to the irradiation position by an air-driven pneumatic transport system into the core of the reactor where the sample will reside until it has been subjected to the specified amount of radiation. Air system operation pressure is 2 bare; the pneumatic transportation system is designed to be operated with compressed air. The air storage tank pressure approximately 10 bare. The rabbit is then extracted from the core and

transported back to the laboratory where the samples are removed from the rabbit, and transferred to non-irradiated vials for completion of the analytical process [28]. A high activity detector mounted to investigate a capsule activity arrives to loading and reception station.

After irradiation, the samples were measured individually for their γ - radiation emission for 4 min. approximately. This short irradiation methodology was used for the determination of Al, Cl, Mg, Na, Ti, V and K [69].

Figure (3.5); Fast transport pneumatic tubes (2 tubes available)

3.4.1.4. Long Irradiation

Sample were accurately weighed and recorded, then coat each one with aluminum sheet, labeled and put together inside aluminum can which closed automatically. After that, it is marked according to definite system, and tested at liquid nitrogen before localization it in the irradiation box according to required flux and remove from the irradiation box after the required time of irradiation. After irradiation, the external dose rate is measured at contact. Vials are stored for (21-25) days at special storage place. The vials are picked out of the can to record the dose rate and surface contamination periodically for each sample, then return all samples to the can again. Nearly after 3 weeks open the can and then measure the samples individually by HPGe according to reference tables which recommend the best decay time before the measurements for each isotope. This means that the sample counted many times where each time for

specific isotope. The long irradiation procedure was applied for the determination of Sb, Mn, Ta, As, Ba, Cs, Co, Eu, Fe, Rb, Sc and Zn.

For the measurements of the long-lived radio nuclides there are many manually loaded irradiation boxes for irradiation of samples. The samples requiring longer irradiation times (hours or days), with moderate to high flux densities, are packaged in high-purity quartz vials. As many as 16 samples can be bundled into a watertight metal container called a sample holder, which is manually lowered into the reactor for the specified irradiation time. After irradiation, the sample holder is removed from the core to cool, which allows the short-lived radioactivity to dissipate to safe handling levels. There are many manually loaded sites for irradiation of samples for several hours or day or more days. Some of these sites are for thermal neutron activation and others for epithermal/fast neutron activation analysis.

3.4.1.5. Photon Measurements

The individual gamma emitting nuclides in irradiated gemstones were easily identified and quantified by using a germanium detector. Although the energy resolution is outstanding, because of its low detection efficiency, this instrument may have limited usefulness in a commercial operation. Counting times of (60) minutes were needed. The object is to measure the energy distribution of the incident radiation [33].

The extra lead reduces background from environmental gamma photons associated with building materials and instrument construction materials. In addition, this extra lead can reduce the ‘‘soft’’ cosmic muon components of background. The lead shielding is usually lined internally with cadmium and/or copper to absorb any secondary x-rays and cosmic components [72].

3.4.1.6. Detector characteristics

3.4.1.6. 1. Energy resolution

Energy resolution of the gamma spectrometers is defined as full width at half maximum (FWHM) in kilo electron volts (KeV) for peak of the cobalt-60 at 1.33 MeV or the peak of cesium-137 at 0.662MeV. The resolution (R) of the hyper pure germanium for the peak of cobalt-60 at 1.33 MeV in neutron activation analysis in ETRR-2 is 2.5 KeV.

3.4.1.7. Measurement of Gamma Rays

The measurement system is carried out by using the HPGe spectroscopy system. The instrumentation used to measure gamma rays from radioactive samples generally consists of a semiconductor detector, associated electronics, and a computer-based, multi-channel analyzer (MCA/computer) [(28)].

3.4.1.8. Counting system calibration

The calibration of the gamma spectrometers defines these three relations:

(1) Spectrum channel numbers to energy,

(2) The FWHM of the peak and energy, and

(3) Spectrum count rate and activity in (Bq) or other units.

The data collected are in counts/unit time/channel, to be useful, it needs to be converted to activities. Calibration is the determination of the proportionality factors that relate the measured activity (peak-area in the γ-ray spectrum) to the amounts of the elements present in the sample under experimental conditions.

3.4.1.8.1. Energy Calibration

The energy calibration calculates two sets of parameters: the energy vs. channel number, and the peak shape or FWHM vs. energy. The inputs are spectrum with isolated peaks distributed over the energy range of interest. By choosing four standard sources (Cs-137, Co-60, Na-22, and Ba-133) for energy calibration, because the gamma ray emitted from these sources cover wide range in the spectrum. The four standards sources are placed over the HPGe detector and start counting for 1800 second and then mark the known peaks and

inform the software that this energy is the energy relating to channel number and repeating this steps until finish all peaks. The final step in energy calibration is to save the energy calibration [28].

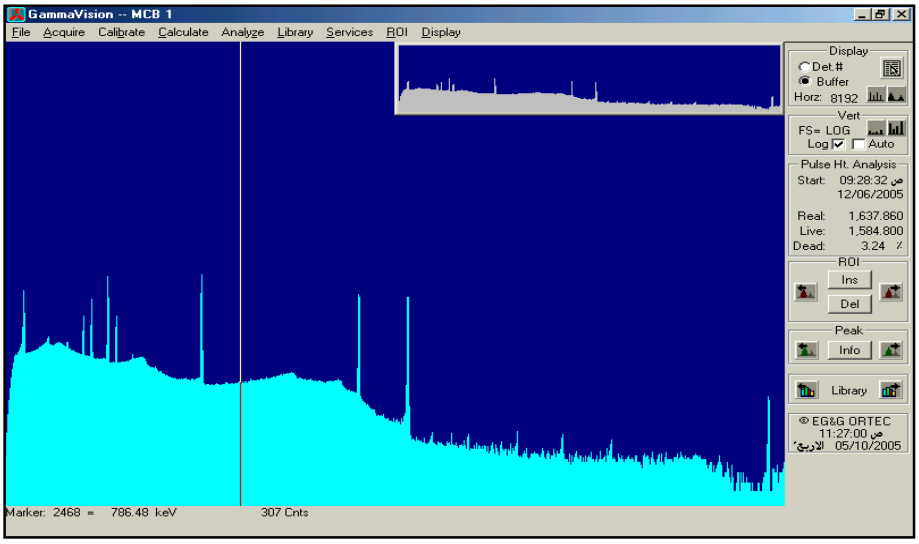

Figure (3.6); Energy calibration spectrum

3.4.1.8.2. Efficiency Calibration

The efficiency calibration calculates the detection of the spectrometers as a function of energy. The efficiency calibration includes effects from the detector itself, the detector-source geometry, the materials surrounding the detector and absorption in the source material or matrix. The efficiency calibration is done by choosing different standard sources to have at least 10 points in the efficiency curve to obtain precise data. The procedure is placing source by source. Then by counting each source separately and mark the peak of interest and calculate the activity of source corrected for decay time and inform the software the activity and calculate the efficiency related to the energy of this peak, and repeat this step until finishing. This gives all sources 10 points in the efficiency curve.

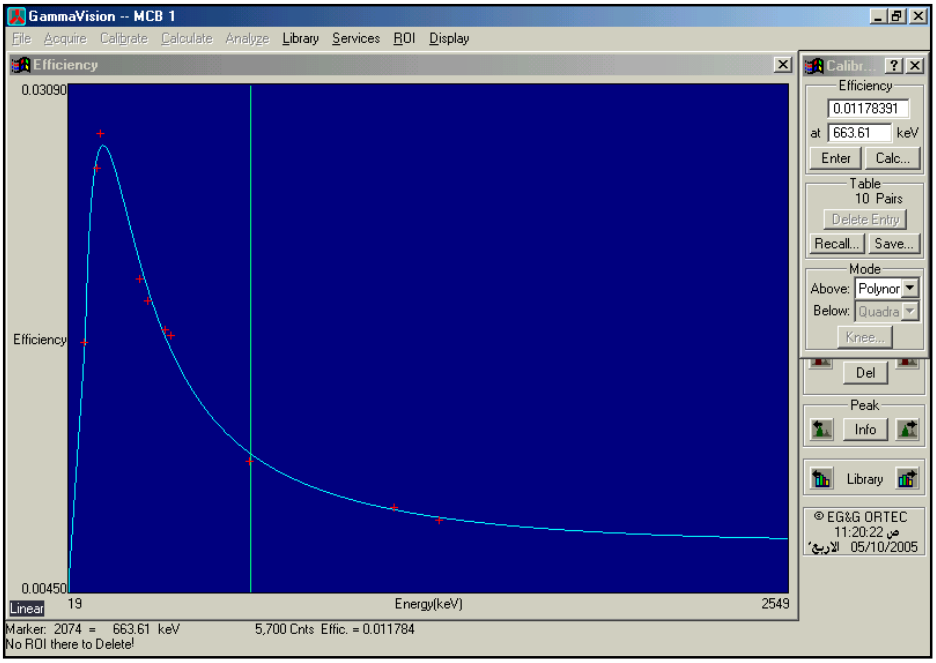

Figure (3.7); Efficiency calibration curve for the HPGe detector

3.5. Raman Spectroscopy [28]

The components used in the raman system, namely, FRA 106/S is including an optional high-sensitivity Ge detector with high sensitivity. A broadband KBr beam splitter with less than 20% signal loss in the raman experiment is used. Cooled Ge detector is required. A complete set of sample holders and test samples are used to check the system.

3.5.1 Raman Spectrometer characterizations:

1. Fluorescence-free Raman spectra with 1064 nm excitation
2. Rapid analysis with a minimum of sample preparation
3. Gold-coated optics for highest throughput
4. Spectral range: -2000 → 4000 cm^{-1}
5. Step scan option for time resolved studies
6. Dual laser/detector option
7. Wide range of sampling accessories

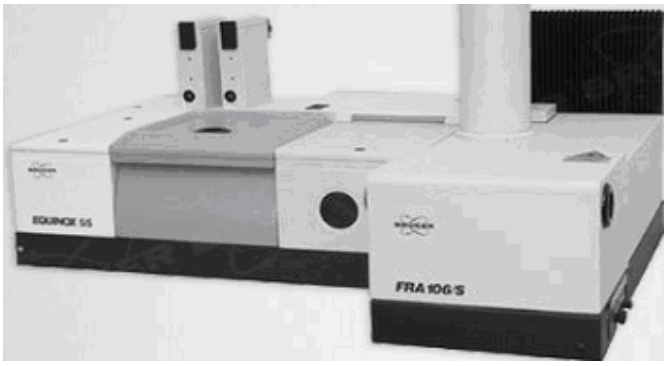

Figure (3.8); FRA 106/S raman spectrometer

3.6. UV/VIS Spectrophotometer

The UV/VIS8500 double-beam spectrophotometer is designed to perform qualitative and quantitative photometric analyses in the ultraviolet and visible regions [73]. Photometric value (% Transmittance or Absorbance) at a single wavelength can be measured. The wavelength slow speed reaches 6,000nm/min. Moreover multi wavelength photometric measurements up to 20 wavelength points can be set up in the multi- wavelength photometric measurements mode. Wavelength scanning is possible with selection of steps of 0.1, 0.2, 0.5, 1.0 and 4.0nm which corresponding to speed ranges from 10nm/min to 3,200nm/min. Light source switching position can be user defined from 320nm. Light source and filters are automatically changed during scanning. Peaks and valleys will be automatically detected after scanning. The UV/VIS8500 allows recording the absorbance or transmittance value of a sample as a function of time at a specified wavelength.

Figure (3.9); UV/VIS8500 Double-Beam spectrophotometer

All specifications and technical data of spectrophotometer used are described briefly in the following:

Table (3.1); Optical Specifications and Physical Characteristics of the Spectrophotometer

Optical Specifications	
Monochromator	Dual-beam, single monochromator grating system 1200 lines/mm
Wavelength Range	190-1100nm
Wavelength Accuracy	±0.5nm
Wavelength Repeatability	±0.3nm
Wavelength Display Resolution	0.1nm
Photometric Range	Absorbance: -0.3-3.0A
	Transmittance: 0-200%T
	Concentration: 0-9999Conc
Photometric Accuracy	±0.5%T
Photometric Repeatability	±0.3%T
Stray Light	≤ 0.05%T@220nm & 340nm
Bandwidth	1.8nm
Stability	< 0.002A/h@500nm
Baseline Flatness	±0.004A
Scanning Speed	Hi, Med, Low, Max: 800nm/min
Data Output	USB standard interface
Light Source	Tungsten Halogen/Deuterium Lamp
Physical Characteristics	
Standard Sample Holder	4-position Cell Holder installed
Printer Interface	HP & EPSON Printers
Power Requirements	100-120 or 220-240 VAC, 50/60Hz
Dimensions	640(L) x 450(W) x 260(H) mm
Weight	30kg

3.7. Positron annihilation:

3.7.1. Positron Lifetime Spectroscopy

Positron annihilation spectroscopy (PAS) is one of the nuclear techniques used in the field of material science. The positron lifetime t is a function of the electron density at the annihilation site. The present measurements are used to study the behavior of defect concentration in one of the most important materials; topaz in two states; pure and irradiated topaz. It has been shown that positrons can become trapped in imperfect locations in topaz samples and their mean lifetime can be influenced by changes in the concentration of such defects. The mean lifetime and trapping rates were studied for the samples.

Figure (3.10); PAS setup

3.7.2. The sample

Natural topaz crystals were cut into smaller samples. Raw materials used were cut using diamond knife, carved, rubbed and polished in the ring face with equal sizes with perfect dimensions for all samples 1 x 1 x 0.2 cm. Then it was treated by different ways. Two identical samples sandwiched the source and all cover with aluminum sheet carefully and directed between two detectors as in the fig. (2.4).

3.7.3. Measurements

The positron lifetime measurements were performed by using fast-fast coincidence system with resolution of 240 ps. The 20 micro-Couri (7.4×10^5 Bq) ^{22}Na positron source inside Al foils, was sandwiched between two identical pieces of the sample under consideration.

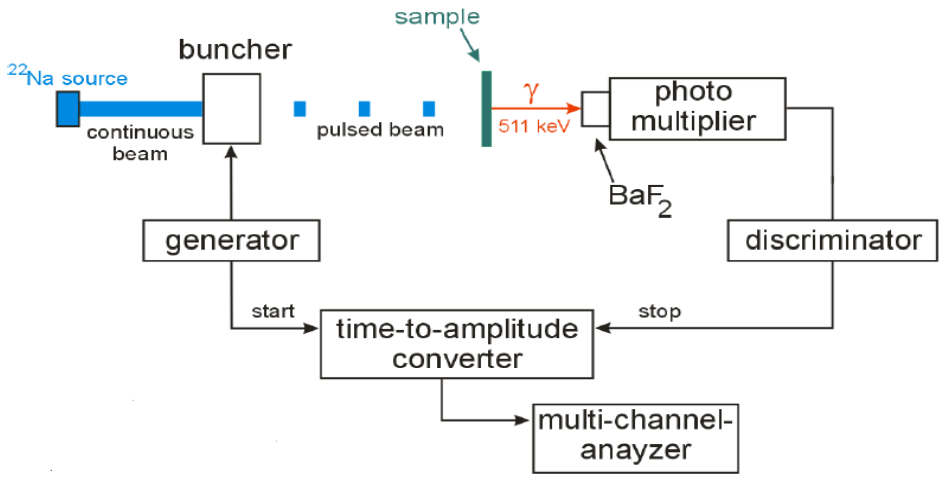

Figure (3.11); PAS using ^{22}Na Isotope Sources

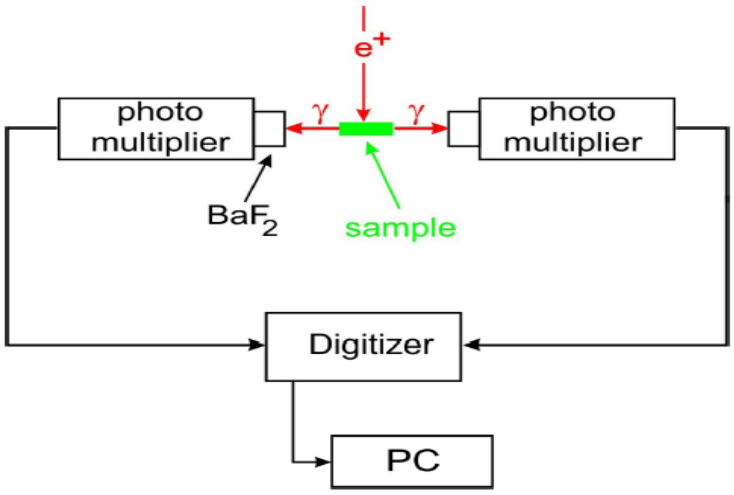

Figure (3.12); Digital lifetime measurement

The conventional positron lifetime measurement is possible since a γ-quantum with energy of 1.27 MeV is emitted almost simultaneously with the positron in the ^{22}Na source. The positron energy, which extends up to 540 keV, decreases in the sample within a few picoseconds by non-elastic interactions. The mean

positron penetration depth of this so-called thermalization process is of the order of 100 μm. The thermalization time usually amounts to a few picoseconds. It is thus small compared with the positron lifetime and can be neglected. On reaching thermal energies, the positron diffuses in the periodic lattice potential before it is possibly trapped in a lattice defect. The diffusion length is in the order of 100 nm. This distance determines the number of atoms to be probed for positron traps during the positron lifetime. Hence, the diffusion length strongly determines the sensitivity of the positron methods to detect defects. The positron lifetime of a single event can be measured by detecting the time difference between the birth γ-quantum of the β^+ -decay in the source and one of the annihilation γ-quanta of energy of 511 keV. The activity of the source must be sufficiently low in order to ensure that on average only one positron is in the sample. This avoids the intermixing of start and stop quanta originating from different annihilation events. A special "sandwich" arrangement of foil source, samples, and detectors guarantees that all positrons emitted from the source are penetrating the sample material. The γ-rays are converted by scintillator–photomultiplier detectors into analog electrical pulses. The pulses are processed by discriminators. Their output pulses start and stop a time-to-amplitude converter as an "electronic stopwatch". The amplitude of the output pulse is proportional to the time difference between the birth and the annihilation γ-quanta and, thus, represents a measure of the positron lifetime. The single annihilation event is stored after analog–digital conversion in the memory of a multi-channel analyzer. The channel numbers represent the time scale. In order to obtain the complete lifetime spectrum, more than 10^6 annihilation events must be recorded. The scheme of the positron lifetime measurement is shown in fig. (3-13) BaF_2 or plastic scintillators and photomultipliers with a short pulse rise-time are used to obtain a high time resolution. The discriminators suppress noise and generate standard timing pulses by the constant-fraction discrimination principle. This principle is

favored over leading-edge discrimination in order to ensure stable time markers independent of the pulse height. Another task is to guarantee that the 1.27-MeV and 0.51-MeV quanta are accepted only in the appropriate channels. The discriminators are of differential type (single-channel analyzer) and accept input pulses within an adjustable energy window.

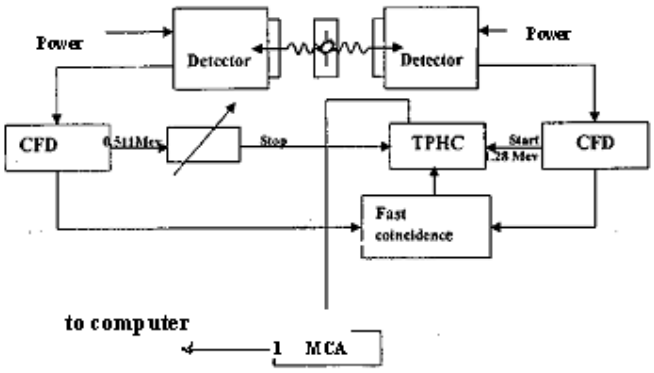

Figure (3.13); Schematic diagram of (PAL) spectrometer

The timing pulses are used to start and stop the charging of a capacitor in the time-to-amplitude converter (TAC). The time linearity is ensured there by constant-current charging that is stopped at the arrival of the stop pulse originating from the annihilation γ-quantum. The stop pulse is coax-cable delayed in order to shift the time spectrum into a linear region of the TAC. The spectrum is stored in a multi-channel analyzer. This experimental arrangement is called "fast–fast coincidence" setup. The term is related to the fact that the time measurement as well as the energy selection is performed in a fast channel. A slow channel was used for energy selection when fast differential discriminators were not available at the beginning of positron lifetime experiments. This arrangement is called a fast–slow setup. Inexpensive multi-channel plug-in boards for personal computers with about 2000 channels are sufficient for storing the spectra. The time resolution of the spectrometer is determined mainly

by the scintillator–multiplier part and ranges between 180 and 280 ps. The practical consequence of this relatively poor resolution is the limitation of the determination of positron lifetime components larger than about 50 ps. The determination of positron lifetimes can, however, be carried out with an accuracy of nearly "1" ps.

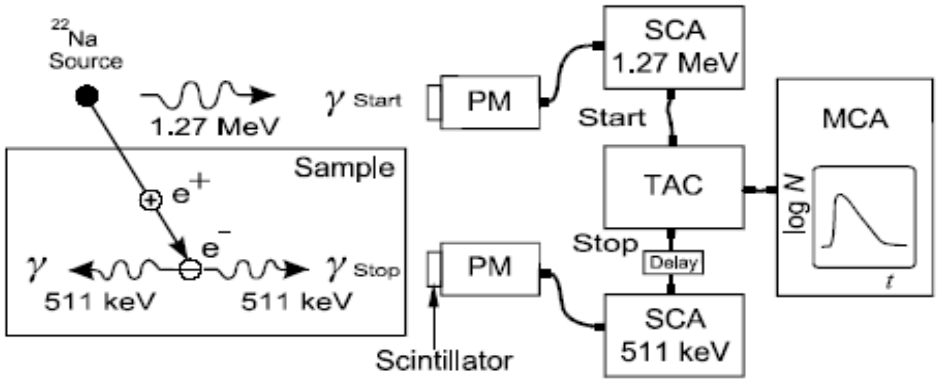

Figure (3.14); Scheme of the positron lifetime experiment in fast–fast coincidence

The lifetime is measured as the time difference between the appearance of the start and stop γ-quanta (PM—photomultiplier, SCA—single-channel analyzer). The amplitude of the time-to amplitude converter (TAC) analog output pulse is proportional to this time difference. The whole lifetime spectrum N(t) is stored in a multi-channel analyzer (MCA) as shown in fig (3.16).

Standard computer programs based on Gauss–Newton non-linear fitting routines are available for the decomposition of the spectra. A model spectrum with a given number of decay components and a given resolution function is used for the least squares fit to the measured spectrum by the variation of the parameters of the lifetime components. The linear parameters, i.e. the intensities and the background, are fitted independently of the non-linear ones, which are the lifetime's t_i and the time-zero channel t_0. The source correction has to be

performed after background subtraction. This means subtracting the characteristic lifetime spectrum of the source. The determination of this source spectrum is rather complicated and usually carried out in such a way that a one-component spectrum of a defect-free sample is analyzed. The source components are varied using a single-component fit to get the best fit, i.e. the smallest variance. The problem of the source correction was investigated in detail on measured and simulated spectra [Staab et al. (1996)] [(75)]. It was found that sources prepared from Al foils provide a three-component source spectrum. The fraction of the annihilations in the source is not only determined by the source itself but it is also a function of the atomic number of the sample and increases due to multiple positrons backscattering through the source. At least *five million events* should be collected for a reliable lifetime separation, even if only two components are present. If only the average positron lifetime is to be measured, a lower number of counts are sufficient.

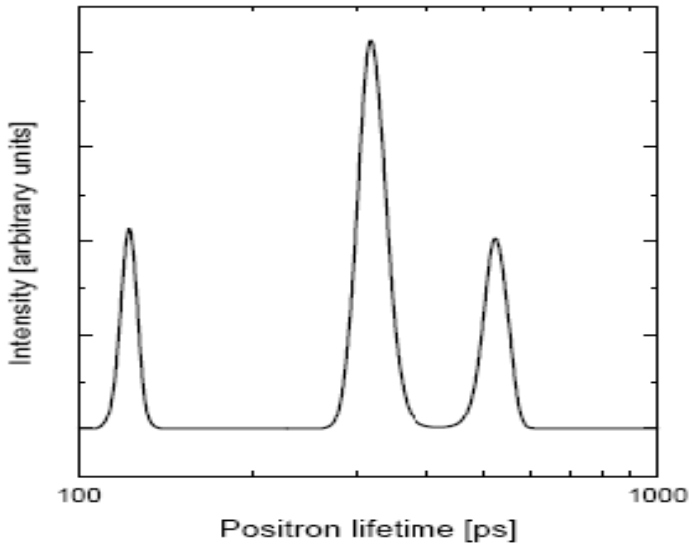

Figure (3.15); PAL spectrum

The output is a graph displaying the intensity versus lifetime. Knowledge of the number of components is not required. This is an advantage for multi-component spectra.

3.7.4. Experimental Procedures

The Fast-Fast coincidence technique, which is described elsewhere [42and76], was used in this study. The resolution function of the system could be measured by using the coincidence as given by the ^{60}Co source. Because there is no time difference (simultaneous) between the two γ-rays emitted from the decay of ^{60}Co, it is preferable for measuring the resolution of the system. Such a resolution function describes the quality of the lifetime measuring system. This is normally described by the full width at half maximum (FWHM) of the prompt curve which is created by the Coincidence between the start and stop signals at a fixed time delay. The prompt curve can be seen as a Gaussian distribution.

The positron lifetime measurement was done by the fast-fast coincidence technique with a FWHM time resolution of 290 ps. The 1274 keV γ-rays were taken as the start signal for TPHC or the "time to pulse height converter" (TPHC), while one of the 511 keV annihilation quanta, resulting from the two photons annihilation in the material, was chosen as the stop signal. The signal from the TPHC or the "time to amplitude converter" (TAG) was fed to the multi-channels analyzer (MCA) for storage. The 1274 keV signal indicates the birth of the positron and the 511 keV annihilation energy indicates the positron death. Thus, the time interval between the above two γ-rays is the lifetime of the positron in the material which measured.

3.8. (Part-4) Radiation protection Instrumentations

3.8.1. Introduction:

Irradiation is used for many materials. Treating gemstones, for example, with irradiation enhances their color [77]. The safety of personnel against ionizing radiation emitted from gemstones previously exposed to neutrons could be achieved by using suitable tools. It is necessary to use suitable radiation measuring devices such as hand-held or portable survey meters and fixed instruments. These instruments are essential for determination of radiation dose, samples analysis and concentrations estimation for isotopes. Industrial uses of radioisotopes include instrumentation and measuring devices (both fixed and portable) [77].

3.8.2. Radiation Detection Equipments

1. The radiation measuring devices in use in the present study are grouped according to type of exposure:

a- Radiation Worker

b- Member of the public.

a) For radiation worker exposure and occupational control

1. Radiation measuring devices
2. Radioactivity concentration measuring devices
3. Area monitoring devices in the work place
4. Personnel monitoring devices in the work place
5. Measurement during storage of the irradiated gemstones
6. Radiation measurements during transport of gemstones containers by air

b) For members of the public

A thorough understanding of radiation detection equipment and information on radiation exposure is necessary to estimate the health risk from irradiated

blue topaz. Domestic processors will need to understand the equipment and its limitations to assess quantities such as exempt release concentration levels [9].

3.8.3. Radioprotection Instrumentations

Table (3.2); Types of used Radioprotection Instrumentations

Equipment	Main Function	Uses
Portable Beta/gamma survey meter	Measuring the photon equivalent dose rate.	For Monitoring of workplace, for storage of irradiated topaz and for transport of irradiated topaz.
Portable Contamination Monitors	Detection of α, β and γ surface contamination.	For storage of irradiated topaz and for transport of irradiated topaz.
Portable Personnel Monitoring	Personnel Monitoring	For Monitoring of workplace.
Fixed Area Monitoring System (SIMA)	Supervise the radiation levels in the work place.	For Monitoring of workplace.
HPGe Spectrometry System (GSS)	Collection & analysis spectra of radioactive samples.	For storage of irradiated topaz and for transport of irradiated topaz.
Hand, Feet and Clothes Monitors (HFCM)	Simultaneous measurement of hands, feet and clothes.	For Monitoring of workplace.

3.8.3.1. Portable Beta/gamma survey meter (P β/γ SM, PTPSM):

The Geiger counter is the most common instrument used to detect radioactivity. However, it only indicates whether a gem or an article of jewelry is radioactive; it cannot determine the type or amount of radionuclides causing the radioactivity [64].

Two distinct radiameters conform the beta/gamma survey meter system

3.8.3.1.1. A Portable Radiameter [78]

This portable dose rate monitor is used for measuring the photon equivalent dose rate (H_x). It uses a proportional counter tube as radiation detector. The default mode is rate meter mode although it can also be used in counter mode. In counter mode, the impulses that are triggered inside a previously selected measuring time are counted. The dose rate is calculated from these values, and displayed. Dose rate, dose rate mean value, maximum dose rate, and total dose are calculated and can be displayed. The determined measured values can be stored in the unit and be transferred to a PC for further processing.

Figure (3.16); Portable Radiameter

Table (3.3); Technical data [79]

1	Measuring range	100 nSv/h (natural background radiation) to 1 Sv/h.
2	Energy range	36 keV - 1.3 MeV
3	Detector sensitivity	2.0 Imp/s per µSv/h

3.8.3.1.2. The Portable Telescopic Probe [80]

Which allows the monitoring of gamma dose rates and beta contamination for external irradiation areas where a changeable distance is possible, thus, reducing radiation exposure hazards. It is attached to the portable radiameter and acts as an external detector to it. The detector of the teleprobe contains two energy compensated (Geiger-Müller) tubes. The alarm levels for this detector can be set independently form the alarms of the portable radiameter. The acoustic alarm is activated upon reaching either alarm level. The probes feature a telescopic extension of more than 3 meters length. This allows for a simultaneous dose and dose rate measurement at both the operator end and the telescopic probe end. It is battery powered, rugged and easy to operate.

Figure (3.17): Portable Telescopic Probes

The teleprobe technical data are:

1. Dose rate measuring range: between 100 nSv/h to 10 Sv/h.
2. It has a built-in speaker.
3. It can log up to 256 measurements for later retrieval by a PC via an infrared serial interface.

3.8.3.2. Large Area Contamination Surface Monitors (LACSM) [81, 82]

These monitors are used for the detection of alpha, beta and gamma surface contamination. They consist of a portable battery-operated measuring unit to which several detectors for different measuring purposes can be attached. They are quick, easy to use and have an extensive memory for readings in isotopes. The detectors used are large area gas proportional counters (either sealed xenon or butane flow counter tubes). A microprocessor provides all functions and calculations.

Figure (3.18): Large Area Contamination Monitors (LACSM)

The importance of monitoring surface contamination in the workplace was first established in the nuclear industry as part of the radiation protection program (Dunster, 1962). A contamination control program is an important component of any radiation safety program. Doses to workers and members of the public arising from contamination in the workplace – and contamination carried out of the workplace – must be maintained ALARA. An effective contamination control program is highly dependant on an effective contamination monitoring program. Surface contamination levels on working surfaces and equipment, protective clothing, personnel, items leaving the working areas and the surfaces in neighboring areas must all be monitored [(83)]. Monitoring surface contamination is considered a necessary control measure for radioactive materials to assess the effectiveness of decontamination measures and the spread of contamination in the work environment [(84)]. The various approaches used to determine the significance of the dermal exposure route have recently been reviewed (Fenske, 1993; McArthur, 1992) [(85)]. Although qualitative information may suffice for assessing the presence of contaminants and the relative hazards in different work areas for risk assessment, quantitative information is required prior to the development of guidelines for maximum

surface contamination limits (Fenske and van Hemmen, 1994) [86]. Thus, it is essential to identify and develop techniques which can be used to determine quantitatively the level of contaminants on workplace surfaces, objectives under study and skin in order to assess a dermal exposure risk or an actual dermal exposure. The development of methods for sampling contaminants on surfaces and the assessment of direct monitoring methods were considered crucial for the assessment of the potential for dermal exposure from occupational surfaces [87].

3.8.3.3. Portable Personnel Monitoring [Mini Dosimeters and (TLD)]:

3.8.3.3.1. Portable Personnel Monitoring [88]:

Electronic dosimeters have advanced rapidly in recent years. They are in widespread use, particularly in nuclear power-plant, home-security, and military applications [34]. Portable personnel monitors are used to determine the dose received by workers or visitors at nuclear plants. Mini dosimeters are pocket size radiation instruments for radiation detection, gamma dose rate measurements and area monitoring. All essential functions can be easily accessed even while wearing protective gloves. The alarm-LED can be seen while the instrument is worn in a belt-holster. The instrument is also equipped with a built-in vibrator and an earphone-output or silent alarming or use in very noisy environment. The mini dosimeters are rugged and reliable and have a removable rubber sleeve for extra protection. They have a large display for clear information with the following characteristics:

1. Radiation type: Gamma and X-ray radiation.
2. Detector: Energy compensated GM-tubes.
3. Measuring range: 0.05 µSv/h – 50 mSv/h.
4. Over range Indication: 1000 R/h (10 Sv/h).
5. Energy Range (+/- 30 %): 45 keV - 1.3 MeV.

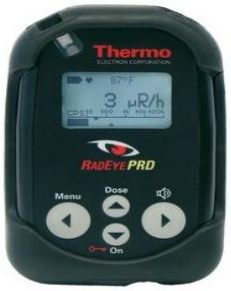

Figure (3.19); Portable Personnel Monitoring

3.8.3.3.2. *Thermo luminescent Dosimeters (TLD)* [90]*:*

It is used to monitor external exposure from ionizing radiation. When heated, the emitted luminescence signal is directly proportional to the radiation dose [91]. These badges should be worn by personnel while on duty. The badge holder is constructed in durable acrylic and carries a thermo TLD-100 chip [91]. These chips must be calibrated. The purpose of calibrating TLD cards is to ensure that all cards in a system will give virtually the same response to a given radiation exposure [91].

Fig (3.20); Portable Personnel Monitoring (TLD) chip and holders

It has the general features such as:

* Reusable hundreds of times.
* Independent of dose rate up to 1000 MGy/s.
* Size: 3.2 mm x 3.2 mm x 0.89 mm.
* Material: Lithium Fluoride.
* Useful Range: 10mGy-10 Gy
* Fading at 20^0C: 5%/yr.

3.8.3.4. Fixed Area Monitoring System (SIMA) [92]:

It is designed for radiological surveillance of any areas where radiation may be present, providing adequate warnings to the personnel potentially exposed to excessive radiation levels. SIMA main function is to supervise the radiation levels in the work places for investigation of dose rate level. It is intelligent system for area monitoring. Each module contains its microcontroller, detector, electronics and low and high voltage supply. Two different alarm levels can be set for each detector.

Each detection module includes a Geiger detector for gamma radiation detection. The digital outputs are used to drive the LCD (Liquid Crystal Display) display, the three LED (light-emitting diode) indicators and the buzzer during testing operation. The digital inputs are used for the counter and to test the state of the push-button.

Figure (3.21); (Area Monitor unit) UMA unit

Detectors are (2) types one for Low and medium dose rate measuring and the other for high dose rate. UMA has four (4) main operating conditions or modes; start-Up, stand-By, operation and shutdown.

The environmental dose rate measuring stages are; reading, analysis, storage, calculation of dose rate and monitor Status. Software of these units is installed on PC and it shows a simplified diagram, which includes, for each UMA; position, identification, actual dose rate and alarm status.

3.8.3.5. HPGe Spectrometry System (GSS) [93:98]:

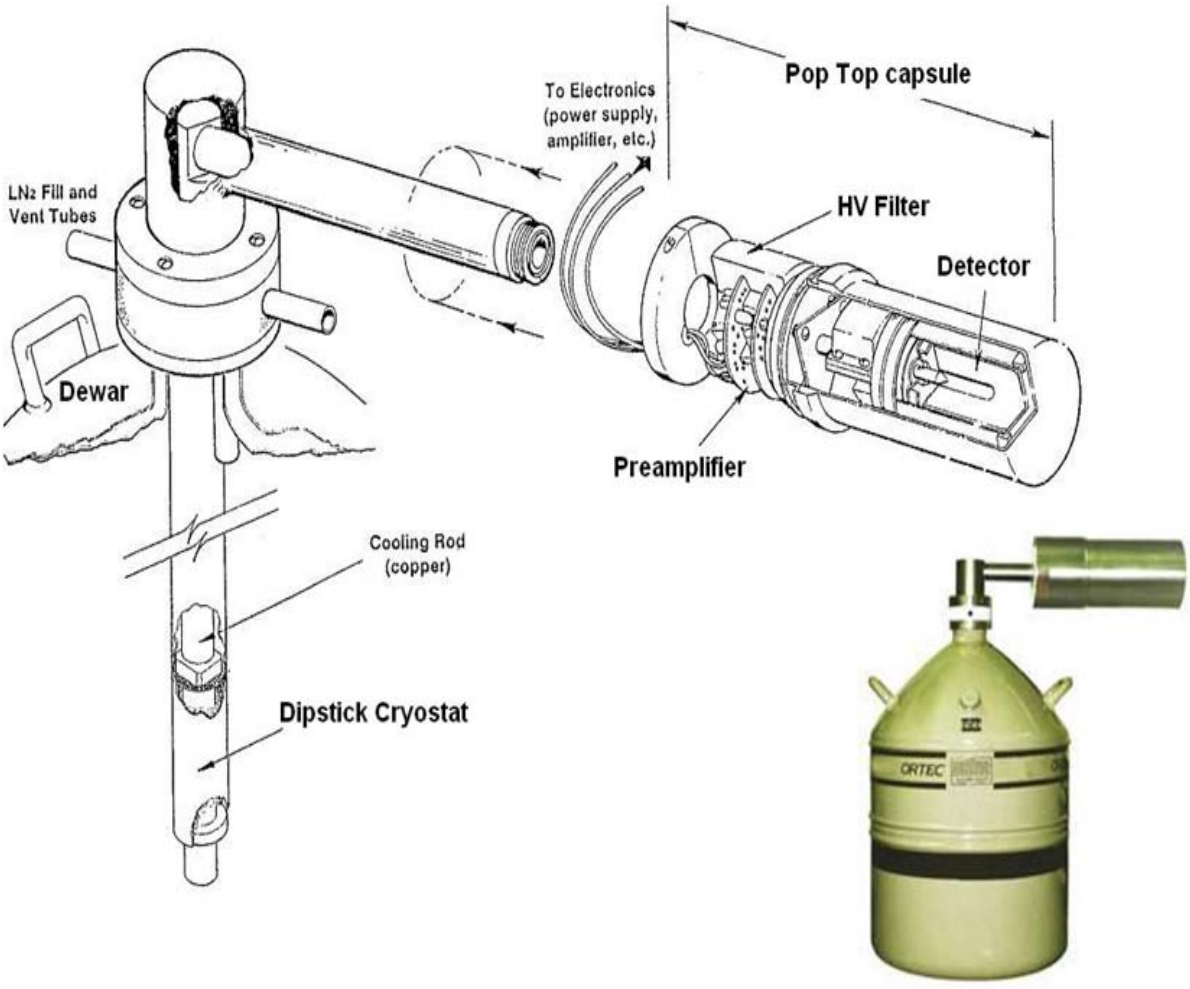

Figure (3.22); HPGe system

Gamma-ray spectroscopy is used in many fields to determine the kinds and quantities of radioactive nuclides induced in various materials. It is now being used more often in gemology to perform research on gemstones and to test them for radioactivity. This includes gemstones like zircons, which contain the natural radioactive elements uranium and thorium [31, 99], as well as neutron-irradiated topaz, which contains radionuclides such as Ta-182 and Sc-46 (Cozar, 1989) [100]. The analytical process is the same whether the radioactivity is natural or laboratory induced.

Gamma-ray spectroscopy is a viable analytical tool because each radioactive nuclide has a unique radiation signature that separates it from all other radionuclides. The radiation (gamma rays) emitted, and the energy or energy distribution of that radiation, identifies each nuclide or isotope present [70]. With a (HPGe) spectroscopy system, just about any gamma ray-emitting radioactive nuclide can be readily identified and quantified [66].

3.8.3.5.1. System components

This Spectrometry system is based on a (HPGe) and consists of:

1. A p-type high purity germanium coaxial detector.
2. A pop top capsule containing the detector, preamplifier, and "HV" filter.
3. A dipstick cryostat which dips into the liquid nitrogen dewar.
4. A 30-liter dewar vacuum container for liquid nitrogen.
5. Digital gamma-ray spectrometer and a detector interface module.
6. A personal computer with maestro-32 MCA emulation software.

Table (3.4); Main characteristics of the detector

Efficiency	10%
Resolution	@122 keV: 0.80 keV @1.33 MeV: 1.75 keV
Peak-to-Compton Ratio	41:1
Peak Shape	FW.1M/ FWHM: 1.9 FW.02M/FWHM: 2.6

The system components are detailed as follow:

The Pop top capsule is the core of the system because the detector element and the front end of the preamplifier are contained in this sealed and self-pumping cryogenic enclosure. The rest of the preamplifier and a HV filter, electrically connected by vacuum feedthroughs, are also part of the capsule. Preamplifiers are used for achieving ultra-high count rates. A temperature-sensing element, also located in the cryogenic vacuum enclosure, automatically

shuts off the HV power supply if the detector temperature begins to rise. If the system is warmed up with bias on, the automatic HV shutdown feature protects the input FET and the critical detector surfaces. The Poptop capsule is cooled by the attached cryostat which in time is cooled by liquid nitrogen contained in the dewar.

A 30-liter Dewar vacuum container for liquid Nitrogen was chosen for the HPGe Spectrometry System. The front-end electronics include the input field-effect transistor (FET) and the feedback element of the charge-sensitive preamplifier. The feedback element comprises a capacitor and a resistor (passive feedback) or a transistor (active feedback).

The Digital Interface Module (DIM) supplies HV for the detector and preamplifier power and warm-up protection for the HPGe detector. It is provided with flying leads to connect to the HPGe detector.

Digital Gamma-Ray Spectrometer (The DSPEC) is a digital nuclear multichannel analyzer system with the following features:

1. Zero Dead Time, the spectrum is corrected pulse by pulse for dead time.
2. With a new kind of digital peak detector algorithm the maximum throughput has been increased by removing some of the dead time associated with the process of pulse peak amplitude determination.
3. It uses a single-cable connection to the detector.
4. It provides high speed control via standard USB 2.0 to any windows 2000/XP/ME system.

And its display has 240 x 160 pixel backlit LCD provides status information, instrument ID, bias information, live and real time where no dead time.

The MCA also has internal battery-backed up memory to maintain settings in the event of a power interruption. The software used is MAESTRO-32. It is an advanced MCA Emulator for use with DSPEC for data acquisition, spectrum display, analysis, and MCB control in the personal computer environment.

3.8.3.6. Hand, Feet and Clothes Monitors (HFCM) [101]

The FHT 65 LL/LLX serves to simultaneously measure the surface contamination of the hands including lower arms and the feet. There four counter tubes; two for hands and two for foots. Optionally, the system allows an additional clothes probe to be connected which may be used to monitor the contamination of other parts of the body or of other small parts that are carried along with the person to be monitored.

During one measurement process, both hands and feet are measured separately. The evaluation of the measuring signals is performed by intelligent preamplifiers. The measured values are displayed on the new display unit FHT 6020. This unit features a LC-display with backlighting where the alpha and beta measured values are indicated. Light emitting diodes (LEDs) and a sound generator signalize optically and acoustically when a failure has occurred or a limit value has been exceeded. It could be connect to PC or notebook. This way, any number of nuclides can be calibrated for the monitor.

Figure (3.23); Hand, Feet and Clothes Monitors (HFCM)

Chapter (4)

Experimental Results and Discussion

4.1. Introduction

The main aim of the present chapter is:

1. To report and discuss the experimental and calculated results,

And

2. To discuss the present work results and its applications in the fields of radiation physics and radiation protection.

The experimental arrangements were previously described in chapter three. Selected samples were analyzed by standard gemological and spectroscopic methods. Theoretical calculations were done to predict the time allowed for transportation. Natural unique crystals of topaz were cut into small and polished samples where they are treated by different ways. Irradiation processes were done inside the atomic energy authority facilities, where neutron irradiation at the Egyptian second research reactor at Inshas site and the gamma irradiation in gamma irradiator unit at Nasr city site.

4.2 Measurements and Analysis

Acquisition of trace element data on gemstones is very important because they determine the market value of the gemstones and could be useful in scientific prospection. Measurements are carried on cut topaz samples were prepared from large piece of raw and treated topaz to have the size of (~ 1.0 cm x 1.0 cm x0.2 cm). Measurements and analysis to identify major, minor and trace elements were determined by (ICP-MS) and (NAA) techniques to determine the elemental constituents of neutron irradiated samples, (PAS) for crystal defects detection, spectrophotometer of a double beam to measure light intensity at different wavelengths, raman spectrometer to observe spectral variations before and after treatment and finally radiation protection

apparatuses for activity of isotopes determination and its concentrations calculations.

4.3 Sequence of operation

1. Raw topaz are mined, cleaned, and sorted.
2. Cutting and polishing then classification.
3. Random analysis of samples by (ICP-MS).
4. Irradiation at reactor or ^{60}Co is performed.
5. Topaz is placed at a special storage with certain arrangements where the dose rate measurements are periodically observed for random samples from different locations.
6. Weighing quantities to be ready for packaging are done.
7. Remove hot stones and weigh it as a final sorting and subtract its weight from all package weight.
8. Analysis by NAA technique.
9. Packaging.
10. Marking.
11. Collecting group together in one bag and re-measuring dose rate at contact and at "1" meter distance from all sides, bottom and top. After these measurements it may be necessary to re-arrangement the packages distribution to modify measurements of dose rate.
12. Labeling.
13. Certificates all required formats.
14. Weighing again by specialist from outside.
15. Signatures.
16. QA & QC revision.
17. The adoption of official certificates of radioactivity
18. Transportation.
19. Finished product shipped to outside or to the market.

4.4. Pre irradiation results

This part shows the results of two selected equipments used for the determination of the elemental analysis of natural topaz namely, (ICP-MS) and (EDS). They are used to know the present isotopes in the raw samples for the best choice of the group of the stones that have the shortest half lives of the isotopes in the stones. Natural topaz stone is usually colorless or has unattractive color. Therefore, it is not considered before irradiation treatment to be a gemstone.

4.4.1. (ICP-MS) and (EDS) analyses:

ICP-MS is a powerful technique for trace multi-element and isotopic analysis. It has been applied to a wide range of samples [(102)]. It has become a valuable instrumental technique for trace multi element analysis due to its greater sensitivity compared to other atomic techniques. This techniques provides the possibility to use fast semi-quantitative analysis programs, that gives faster results than quantitative modes which needs series of external standard solutions, more handling time and moreover require higher reagents for consuming [(69)].

The ICP-MS instrument used through this study was the high-resolution JMS-PLASMAX2. Argon gas flows were controlled by mass-flow controllers. Typical operating parameters of the ICP-MS and key method parameters of plasma conditions and data collection were achieved.

4.4.2. EDS

EDS elemental analysis technique was performed and showed that the major elements are (O, F, Al and Si) with concentrations of (35.62, 19.35, 30.76 and 14.27), respectively. The Half-life time of these major elements are (29, 11.4, 134.4 and 9432) Sec, respectively [(28)]. This means that the Half-life time of the major elements in topaz are very short. Therefore, it is expected for samples of this kind to be used as gemstones because they didn't need long time in storage for their residual radioactivity to die. This could be valid in cases where

samples do not have high concentrations of trace elements that having long half-life times. Therefore, these samples could be used in this kind of work.

4.4.3. (ICP-MS) Measurement Results

Table (4-1) shows average concentrations of the trace elements in five selected samples of cloudy white topaz as measured by LA-ICP-MS and expressed in ppm.

Table (4-1) Average trace elemental concentrations in topaz (ppm) as analyzed by (LA-ICP-MS)

Element	ppm	Element	ppm
Sc	2870 ± 84	Nd	8.2 ± 1.2
Cr	15.2 ± 0.5	Sm	2.6 ± 0.8
Mn	63.6 ± 1.5	Gd	138 ± 2.3
Fe	5012 ± 5.9	Dy	1.8 ± 0.1
Zn	13810 ± 566	Ho	0.2 ± 0.07
Rb	26.3 ± 1.3	Er	1.9 ± 0.3
Sr	42.4 ± 2.3	Tm	0.9 ± 0.06
Zr	7608 ± 23.3	Yb	3.7 ± 1.1
Nb	917 ± 33.8	Lu	0.53 ± 0.2
Co	0.07 ± 0.05	U	6.8 ± 0.8
Mo	99.9 ± 4.3	Ta	7 ± 0.3
Ag	7.5 ± 1	La	2.11 ± 0.5
Cd	6.2 ± 0.6	Ce	114.4 ± 0.9
Sb	40.3 ± 0.8	Pr	4.6 ± 0.3
Th	13.2 ± 1.1		

Inspection of table (4-1) shows the elements at topaz sample as detected by (LA-ICP-MS). From the experience it is found that concentrations of five elements namely Sc, Mn, Fe, Co and Ta are the most important factors for the selection of the samples to be irradiated where they are dominant and control the storage time for decay until achieving the safety level for transportation according to IATA regulations. And whenever those elements have low concentrations in the selected samples they are of the best choice.

4.5. Iirradiation

The samples are irradiated for (2,4,8 and 12) hours at 22 MW- with an average of thermal neutron flux of 2.37x 10^{14} n/(cm2.s). After irradiation of the stones, they have to be transferred to the auxiliary pool until their radioactivity reduces to permissible level, and then to hot cells before any treatments.

4.6. Post irradiation Analysis of topaz

4.6.1. Neutron Activation Analysis (NAA):

(NAA) was used for samples analysis after irradiation. The goal here was to obtain a profile of the main impurities existing in the stones of each region. Any radioactive counting analysis begins with eliminating all removable radioactive surface contamination. Therefore, the surface of each gemstone was thoroughly wiped with a damp piece of absorbent filter paper that was subsequently checked for radioactivity in a shielded Geiger tube assembly. The paper registered background only, which showed that the two gems were free of any removable contamination. As a further precaution, the gems were washed in a standard radioactive decontamination soap solution. Prior to analysis, each was weighed on an electronic balance [(66)].

NAA measurements were performed by means of a γ-ray spectrometer made of a HPGe detector after irradiation in the swimming pool nuclear reactor (ETRR-2) at the nuclear research of EAEA, Egypt. NAA measurements for trace element analysis were carried out using long- and short-time irradiation conditions, according to the neutron cross section of the parent nuclides of elements to be determined [(28)].

Measurement of the samples is carried out at a 7 cm away from the top of the high pure germanium detector with relative efficiency of 100%, and a resolution at full width at half maximum (FWHM), namely 2.1 KeV for the 1332.4 KeV photons of Co-60 [(28)].

For the analysis, each stone was placed inside the one-ton lead shield in the same position as the stimulants to which the system had been calibrated. Counting times for (10,000 seconds, or about two-and-three-quarter hours) were established on the basis of the gem's distance above the detector and the gross radiation readings from the Geiger counter.

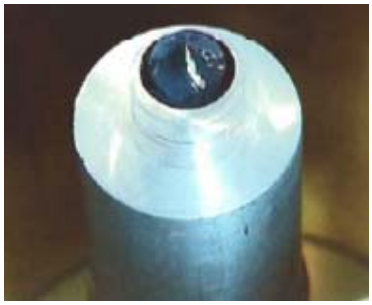

Fig.(4.1); The high-purity germanium crystal detector is housed in an evacuated container that is enclosed in a lead shield and the sample stone has been placed with its center of mass 7 cm above the detector.

Samples are prepared for short irradiation in a polyethylene vials and for long irradiation in high purity grade aluminium foil along with another sample of standard reference material. An empty aluminium foil with the same weight is also included for identifying and subtracting the background of the γ-ray lines due to aluminium envelopes. All samples are enclosed in aluminium cans prepared for irradiation. Irradiated samples are measured after several cooling times.

The gamma-ray spectrum for each sample is collected for different times after many times of cooling.

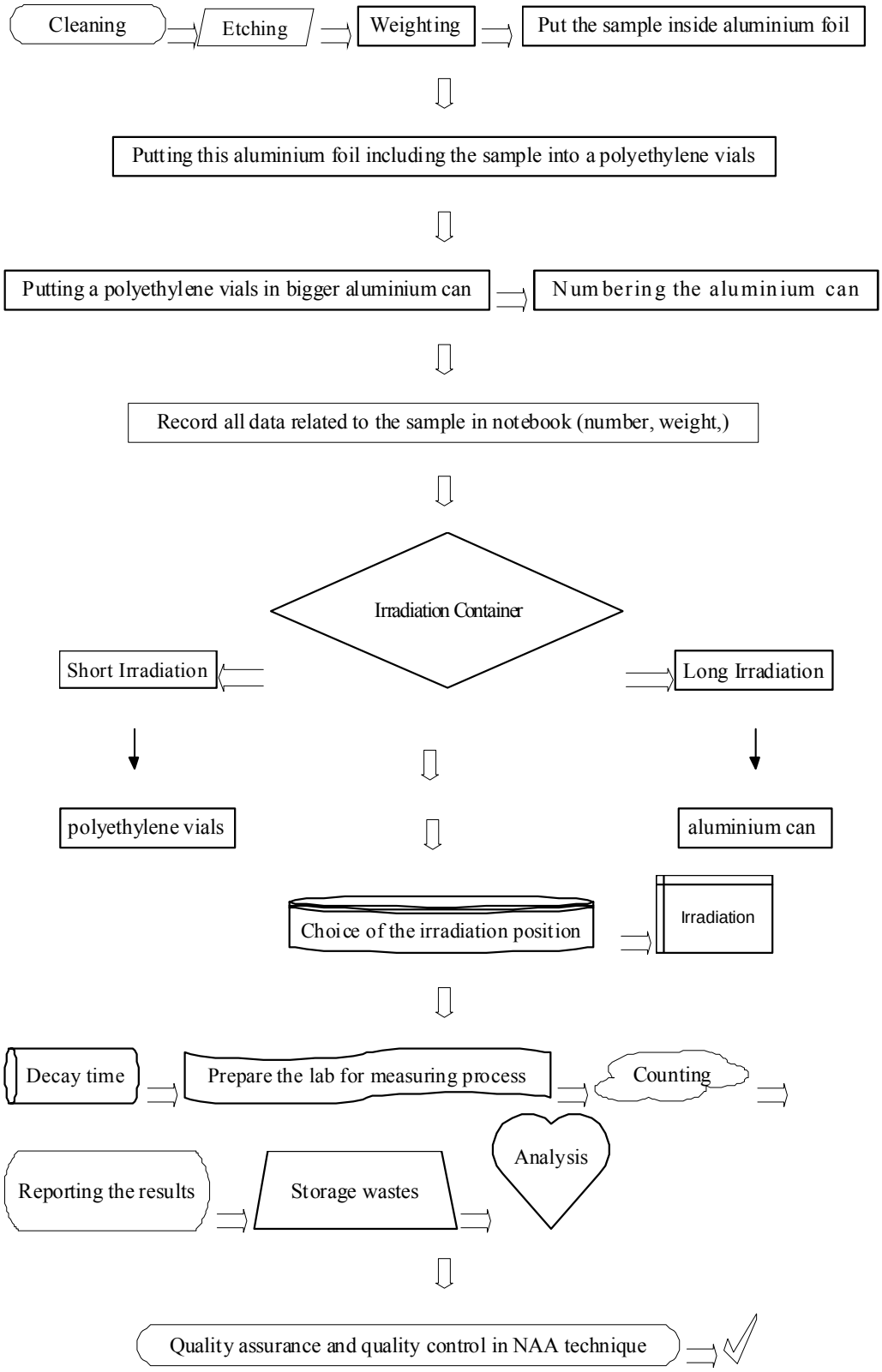

Figure (4.2); Sequences of sample preparation, irradiation, counting and analysis

The irradiation grid in the ETRR-2 reactor includes 26 places as irradiation cells. Two sets of irradiation procedures, namely short and long irradiations are carried out. In the short irradiation the samples and standards are sealed in polyethylene vials. Irradiation for 30 s at 22 MW in the pneumatic transfer tube of ETRR-2 at thermal flux of 6.8 $x10^9$ n/(cm^2.s) is performed. Then samples are left to cool for minute range dependent on the desired isotope count where the gamma spectrum for each sample is collected. In the long irradiation where samples and standards are irradiated for (2,4,8 and 12) hours at 22 MW with an average of thermal neutron flux of 2.37x 10^{14} n/(cm^2.s). Set of topaz samples are irradiated in the same conditions at the same time but for (30) minutes. Different samples from different locations were used in each time. Each time decay of the sample and concentrations of isotopes in the samples were done. The gamma-ray spectrum for each sample is collected for different times after many times of cooling y using the hyper pure germanium detection system.

The emitted gamma rays are identified according to the energies of the well-resolved gamma-ray lines. To satisfy the accuracy of the detection system, the results of the present study of the reference material of some elements which appear clear and well resolved are compared with the certified values of this standard material. Energy calibration curve for HPGe detector gives linear relation between channels vs. energy. Energy calibration peaks by using Co 60 (2 lines at 173.2 and1332.5), Cs 137 at (661.62) and Ba 133 at (302.84, 356, 383.85 and 276.4) KeV. Also efficiency calibration was done after energy calibration and before counting.

4.6.1.1. NAA Calculations

The samples which irradiated and later measured under the same counting conditions were used to calculate the concentration of the elements of interest by comparison of the measured activity between the sample and the standard.

Some residual radioactivity was observed for some samples several times higher than the background even after 6 months of irradiation. This activity is due to the higher concentration (compared with the previous samples) of the elements (Sc, Fe, Mn and Ta) with half-lives of (83.79, 44.4, 312.3 and 113.5) days, respectively.

To calculate the radionuclide content of a gemstone, the analyst must determine the detector efficiency at all gamma-ray energies of interest. The number of gamma rays completely absorbed by the germanium crystal detector depends on the energy of the gamma rays; the size, density, and dimensions of the gemstone being analyzed; and the distance of the gemstone from the detector. Special radioactive calibration sources were made for GIA by Analytics, Inc., of Atlanta, Georgia, from a resin with a specific gravity of 1.15. These sources were constructed to simulate the size and shape of various gemstones, because the closer the stone to be analyzed approaches the size, shape, and counting geometry (position above the detector) of the calibration source, the closer the calculated results are to the true values [66].

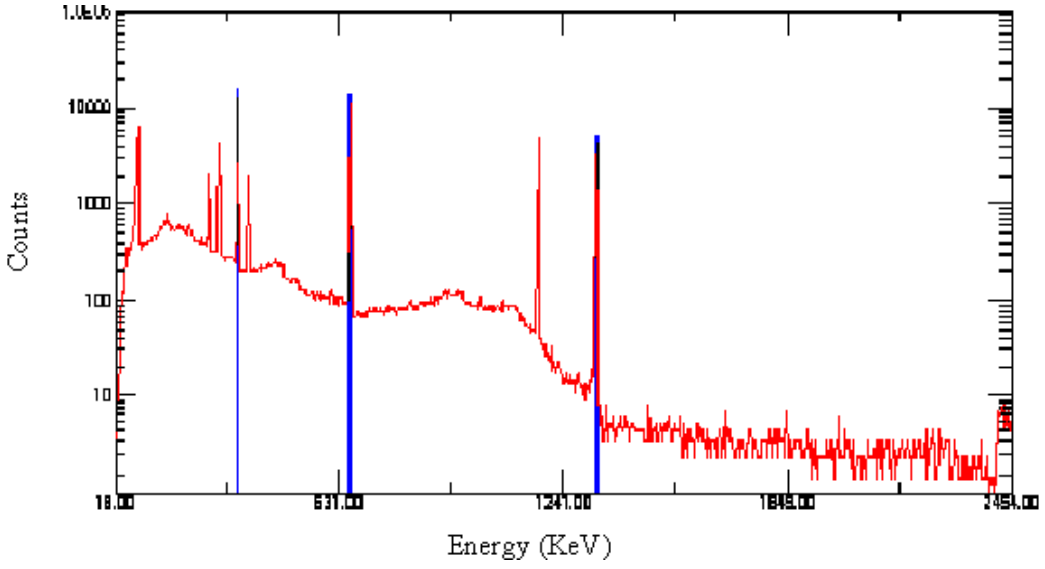

Figure (4.3); Energy Calibration (using Co 60-Cs 137 and Ba 133 isotopes)

4.6.1.1.1. Relative method [28]

This method depends mainly on the ratio between concentration of standard material and its gamma peak intensity, where:

$C_e / C_s = A_e / A_s$ → (4.1)

Where C_e is the concentration of interested element, C_s is the certified concentration of element in the reference material, and A_e and A_s are the specific activities of element in unknown sample and reference material, respectively.

Where: *The specific activity = activity (Bq) / weight (gm)*

The initial activity (A_0) can be calculated as in the following:

$A_0 = A / [(e^{-\lambda td}) (1- e^{-\lambda tc})]$, → (4.2)

A = current activity, $\lambda = \ln 2 / t_{1/2} = 0.693/ t_{1/2}$ And t_d, t_c are the times of decay and counting, respectively.

C_e can be calculated from another relation that depends on the area under the peak.

$C_e = [C_s (A_s / e^{-\lambda tds})]/ (A_e / e^{-\lambda tde})$, $\lambda = \ln 2 / t_{1/2} = 0.693/ t_{1/2}$ where A is the activity and equal to (net area / SDCm), $S = (1- e^{-\lambda ti})$, $D = e^{-\lambda td}$ and $C = (1- e^{-\lambda tc}) / t_c$, t_i = irradiation time ,t_d = decay time and t_c = counting time and me = 2.7084 g , ms = 0.0324 g

4.6.1.2. NAA results and discussion

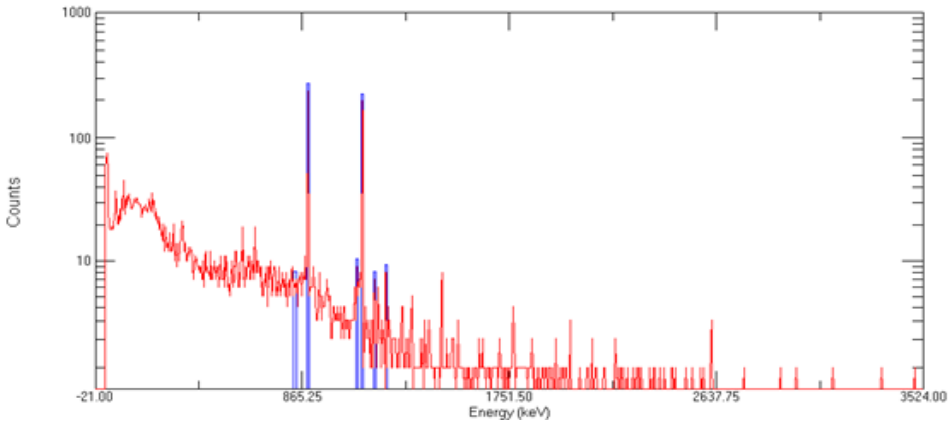

Figure (4.4); γ spectrum of irradiated topaz by neutrons

Topaz was found to contain (Ta-182, Sc-46, Fe-59 and Mn-54), in decreasing order of activity. Nuclear interactions within trace elements that present in topaz produce these radionuclides.

Ta-182 ($t_{1/2}$ =114.43 d) and Sc-46 ($t_{1/2}$ = 83.79 d) are created by neutron absorption reactions from the natural stable isotopes Ta-181 and Sc-45, respectively;

$$^{181}Ta + n \rightarrow {}^{182}Te$$
$$^{45}Sc + n \rightarrow {}^{46}Sc$$

Mn-54 is mostly produced by the absorption of a neutron into iron-54. (Fe-54) which is one of the stable isotopes of iron and that immediate release of a proton [66], Where Fe-59 is coming from Mn-59 by β^--decay. Those are shown in the following equations:

$^{54}Fe + n \rightarrow {}^{54}Mn + p$, where the target is stable and the half-life of the product is 312.3 d.

$^{59}Mn \xrightarrow{\beta^-} {}^{59}Fe$, where the half-life of the parent is 4.6 s and the half-life of the daughter is 44.503 d.

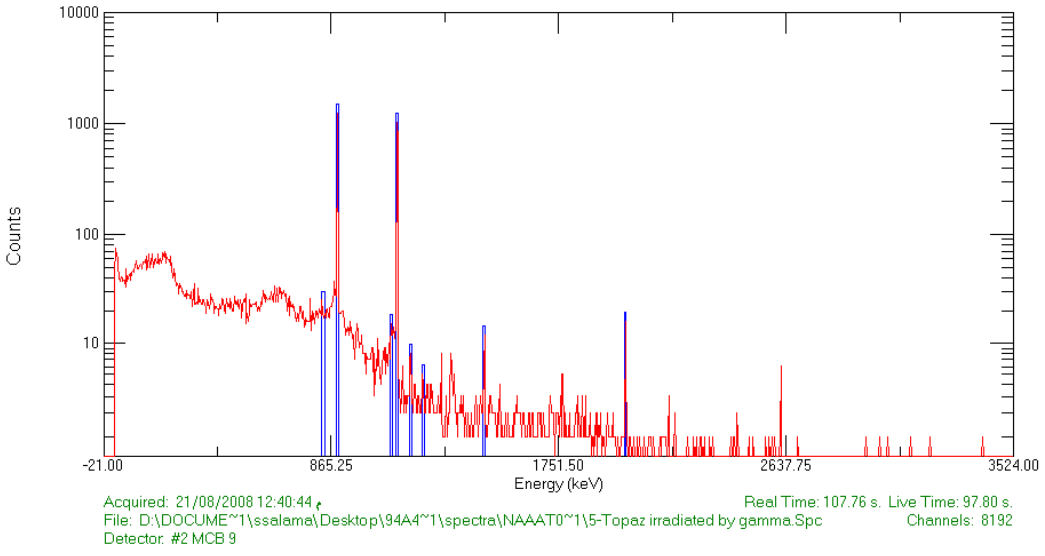

Figure (4.5); γ spectrum of irradiated topaz by neutrons after γ irradiation

As an example irradiated topaz sample, revealed 2.858 nCi/g for Ta-182, 0.858 nCi/g for Sc-46, and 0.238 nCi/g for Mn-54(where 1 nCi=37 Bq). The release date calculated by computer program for decay calculations by sum of the ratios of the radionuclide concentration (i.e., the sum of the concentrations divided by the release-limit concentrations for each radionuclide).

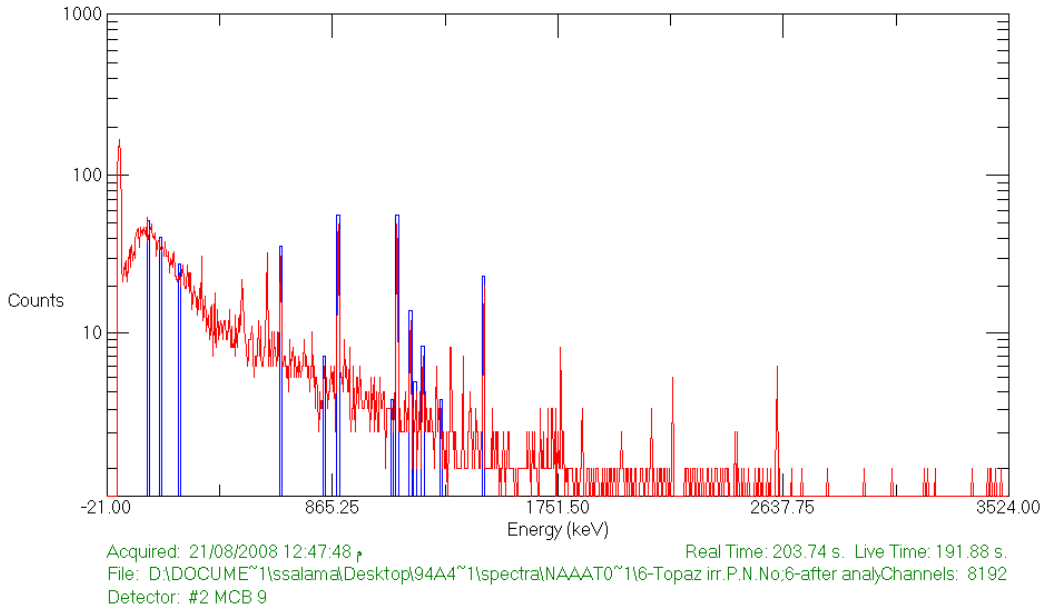

Figure (4.6); γ spectrum of twice irradiated topaz by neutrons

Gamma spectrum of short irradiated topaz shows that it consists of many isotopes with short half-life. Qualitative and quantitative studies by NAA technique for topaz samples showed the following short life isotopes:
[(Al-28), (CL-38), (Rh-106M), (Kr-88), (Bi-214), (Rb-89), (Ac-228) and (As-76)] with specific activities:
[(736345.91), (1.5), (10.6), (22.4), (91.4), (11.4), (153.5) and (41.7) (Bq/g)] for each isotope, respectively [28]. Activities of these isotopes go to vanish during days. The isotopes with short half-life as Al-28(2.24m) and Na-24(15.03 hr) could be detected by the pneumatic transport system. It was noted that the major elements in topaz have short life time. Therefore, topaz is preferred in the gem stone industry to be used as an industrial product from the reactor.

Qualitative and quantitative studies by NAA technique for long irradiated topaz show that it consists of *Sc-46(889.25), Mn-54(834.83), Co-60(1332.5 and 1173.2), Cs-134(604.7and 795.8) and Ta- 182(1121.3,1189.1 and 1221.4)* Bq/g as isotopes mainly producing the residual radioactivity [28]. The isotopes with short half-life times disappeared and the isotopes with long half-life times such as Sc-46 (83.85d) and Ta-182 (114.74 d) started to appear [28].

Table (4.2); Impurities as found by NAA in topaz samples which are irradiated by neutrons

Element	Nuclide	Half-life
Ag	Ag-110	249.76 d
Al	Al-28	2.24 m
As	As-76	26.32 h
Au	Au-98	2.7 d
Br	Br-82	35.3 h
Ce	Ce-141	32.5 d
Cl	Cl-38	37.24 m
Co	Co-60	5.27 y
Cr	Cr-51	27.7 d
Cs	Cs-134	2.062 y
Eu	Eu-152	13.33 y
Fe	Fe-59	44.4 d
Ga	Ga-72	14.1 h
Hf	Hf-181	42.39 d
La	La-140	40.27 h
Mg	Mg-27	9.46 m
Na	Na-24	14.96 h
Rb	Rb-86	18.66 d
Sb	Sb-122	2.7 d
Sc	Sc-46	83.1 d
Ta	Ta-182	114.5 d
Th	Th-233	22.3 m
Zn	Zn-65	243

4.7. Validation of ICP-MS and NAA data

Samples were analyzed by NAA and ICP-MS. A general agreement can be observed when comparing NAA and ICP-MS data. To evaluate of the accuracy of the methods employed, a certified sample was analyzed by both techniques. Regarding the values obtained, it is interesting to point out the general agreement between both techniques in all the samples. However, for Mn, Fe and Co, there are differences between the results obtained by both techniques and this may be due to a poor recovery of the sample digestion, which is necessary for the determination by ICP-MS.

The short time required for the analysis by ICPMS is one important advantage over NAA, especially in the case of long irradiation experiments. There are two additional advantages of ICP-MS over NAA; the lower cost of the analysis and ICP-MS implies smaller negative influence on the environment and reduced health risks for the analyst.

4.8. Raman Spectroscopy Measurements:

The Raman measurements were performed using a backscattering geometry at room temperature in the range from 400 to 4500 cm^{-1}. These measurements were done using Fourier Transform Raman Spectroscopy (FTRS) "FRA 106/S" equipped with an optional high-sensitivity "Ge" detector and a "CaF2" beam splitter.

Raman spectroscopy is used to identify the color centres aiming to understand the processes of color in samples of treated gemstones. The stones were placed in front of laser filters on a suitable low temperature holder blocking all lasers light which was not hitting the stones.

4.8.1. Raman studies on topaz

Topaz was subjected to intensive Raman spectroscopic studies, as a raw material and after treatment by irradiation with neutrons and γ. Spectra of all samples were recorded in the range of 4500–400 cm^{-1} at room temperature.

The effect of the different parameters of irradiation such as irradiation time and position of irradiation were considered. Quantitative and qualitative analysis were carried out to investigate the effect of each parameter and its role with structure changes. Samples used were selected and prepared in suitable manner that they have the minimum contamination of elements rather than the original crystal structure. The absorption bands are correlated with chemical composition and structural parameters.

The application of Raman microprobe is given for identification of natural and irradiated topaz. Raman spectroscopy is used also for investigation of micro-areas inside zoned crystals and to the determination of composition of solid inclusion inside objects. This method is non-destructive, easy as well as fast in use and also allows obtaining enough information about elements composition.

4.8.2. The range of OH group in topaz spectra

Raman studies of interested topaz stones showed a relation between the color changes with changes in the intensities of the band of scattered peaks corresponding to OH group stretching modes of vibrations.

Structural defects result in anomalous weak vibrations in the region between 3000 and 4000 cm^{-1}. Based on the crystal structure, that contains hydroxyl groups showing partial OH-F substitution, it is expected to observe OH-stretching vibrations in this region, although there are contradictions in the exact positions of the OH stretching vibrations where single band is observed around 3143 cm^{-1}.

Typically, colorless or pale-colored topaz is heated to 300 ^0C for several hours. The longer the stone is heated the deeper the color change will occur in the stone. No changes under 200 ^0C were observed. The stones will turn to a yellow to brownish green to a dark brown color above this degree. These colors, however are not stable and will eventually fade to clear unless the stones are irradiated. The irradiation process essentially eliminates the yellow-brown and green colors and leaves a stable blue color which will not fade unless subjected to temperatures of 500 to 600 ^0C at the case of γ irradiation while no effects in neutron irradiation state to highest temperatures.

4.8.3. Stretching vibrations bands

Several authors [28] showed that Raman spectra have the following O…H-hydroxyl group with different stretching vibrations bands:

Al-OH (Bands between 1160 and 1079 cm^{-1}) and F-OH (Bands between 3000 and 4000 cm^{-1}) are taken into consideration [28]. These bands are based on the crystal structure, which contains hydroxyl groups showing partial OH-F substitution. Comparison between pure topaz and irradiated one by neutrons for (2, 4, 8 and 12) hr. by Raman Spectroscopy was done [28]. The Peaks at 156 and

241 were found in natural topaz but they disappeared completely in all times of irradiation. New Peaks at new positions were appeared in the cases of heating, cooling and irradiation for different times. Many peaks have been shifted by different values in all times of irradiation. The position of these peaks did not change in all times of irradiation but their intensity was changed such as one at 268 cm^{-1}. Peaks at 83, 854 and 1008(Irradiation. 2 hr), at 154, 403(irradiation. 2, 8 and 12 hr), at 457(irradiation. 2 and 8 hr), at 924 pure and (irradiation. 2 and 4 hr), at 925(irradiation. 12 hr), and at 935(irradiation. 8 hr) appeared at the previous values. Also bands and its intensity in natural and irradiated topaz sample for {2, 4, 8 and 12} hr were cleared. Besides to band assignments in the Raman spectrum of topaz, compared with (4) different literature data associated with Assignment and its own studies [28].

4.8.4. Topaz spectra at room temperature, heated and cooled

The Raman spectra of topaz shows that there are differences between topaz spectra at room temperature, heated to 240 C^0 and the other one which is cooled to (-100C^0). In the spectrum of topaz sample relatively low intensity bands are observed at 359.368, 457.593, 548.668, 561.2 and 643.3 cm^{-1}. However bands at 854. 98 and 1078.335 cm^{-1} is clearly absent in topaz spectrum after heating and upon cooling to (-100C^0). These bands were present in the room temperature spectrum as very weak bands only. The low frequency region will, in addition to the Al-OH vibrations which are weak, be dominated by Al-(OH) and Al-F vibrations from the $AlO_4(OH,F)_2$ octahedra, internal vibrations from the isolated SiO_4 tetrahedra and Si-O-Al vibrations from the shared oxygen's between the octahedra and tetrahedra. The complexity of the spectrum hindered them to identify the large number of bands in especially the low frequency region. The bands observed around 1005–1008, 982–985, 921–927, 854–855, 559 and 285 cm^{-1} are assigned to the various Si-O modes of the SiO_4 group. The bands observed in the frequency region below 500 cm^{-1} agree with those

observed by *"Beny and Piriou"* [28] on an oriented single crystal. They [28] observed a broad band at 315–335 cm^{-1} which is splitted [28] in two easily recognizable separate bands at 317.59 and 332.921 cm^{-1}. These bands are associated with Al-F stretching modes. The other bands in this low frequency region are probably associated with various Al-O modes.

At room temperature topaz shows two bands around 1078 and 1164 cm^{-1}. These bands are thought to represent the vibrational band (Al-OH) according to *"Wunder and Marler"* [28] ,but *"Beny and Piriou"* reported only one band at 1165 cm^{-1}. They assigned to the in plane bending mode of the hydroxyl group in agreement with the assumption of *"Adams and Hill"* [28]. More than these Bands and its Assignments in topaz sample irradiated for different times and have different values of color (7 colors) were reported [28]. A full comparison for Bands and its Assignments in normal topaz sample and irradiated one for 8 hr (3 Positions) was tabulated also.

The Raman spectrum of topaz shows some bands at different wave numbers. These bands are observed to be changed according to irradiation type. There are some minor differences between topaz spectra at room temperature at different power for Raman spectrometer. Raman spectrum for natural Topaz was taken at different power for the Raman unit (500, 1000 and 1400) Watt. The best results are obtained at 1400 Watt.

The Raman spectra of the colorless topaz from Brazil, as analyzed in this work were comparable to other works [28]. The natural samples or the samples thermally treated display similar spectra. The bands at 646, 847, 934, 983 and 1163 cm^{-1} are related to SiO_4 [28]. The two OH-stretching bands around 3640 and 3650 cm^{-1} present a faint evolution with the thermal treatments; the shoulder at lower frequencies tends to increase as the temperature of the thermal treatments increases [103].

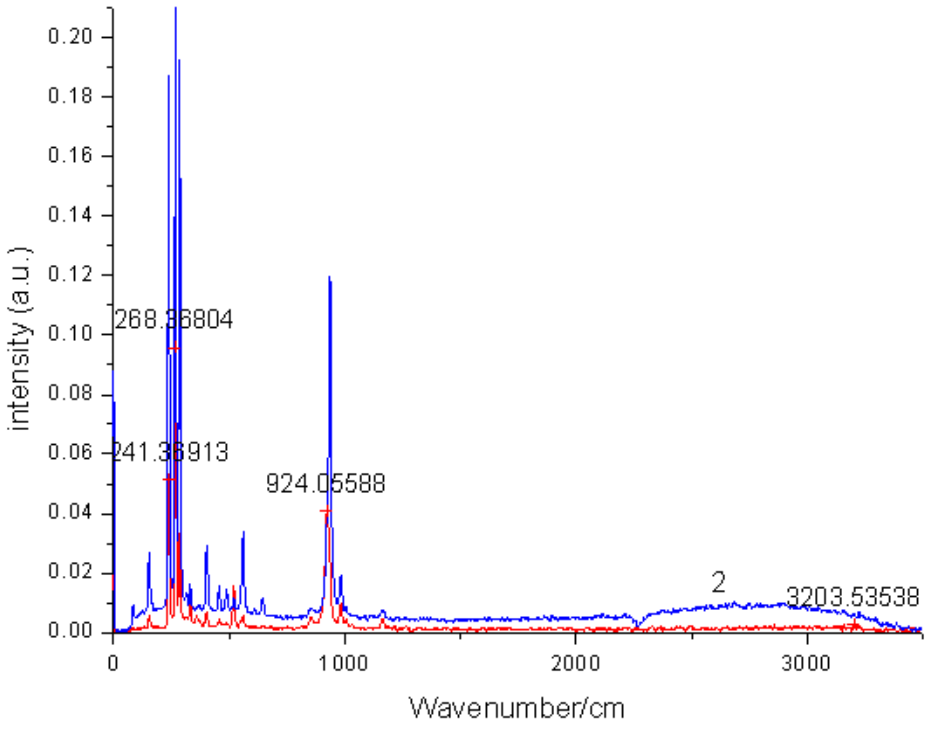

Fig (4.7); Raw Raman spectrum of topaz sample *down* and neutron irradiated topaz *up*

4.8.5. Point defects

Impact of neutron flux in the range of $2x10^{14}$ n /cm^2 /s into topaz can cause point defects [104] due to OH mobility in the crystal structure of topaz. Longer irradiation causes clustering of these defects and more mobility of defects giving rise to a strong optical absorption in the visible region [105]. The band of raman scattered peaks between 3000 cm^{-1} and about 3500 cm^{-1} corresponding to (OH) stretching modes of vibration is recorded.

4.8.6. The effect of the irradiation time on OH group

Inspection of the Raman scattered peaks at about 3000 cm^{-1} show that, by increasing irradiation time and temperature can lead to an increase in the scattering peaks. This may mean that, neutron irradiation helps in band cleavage of the OH group.

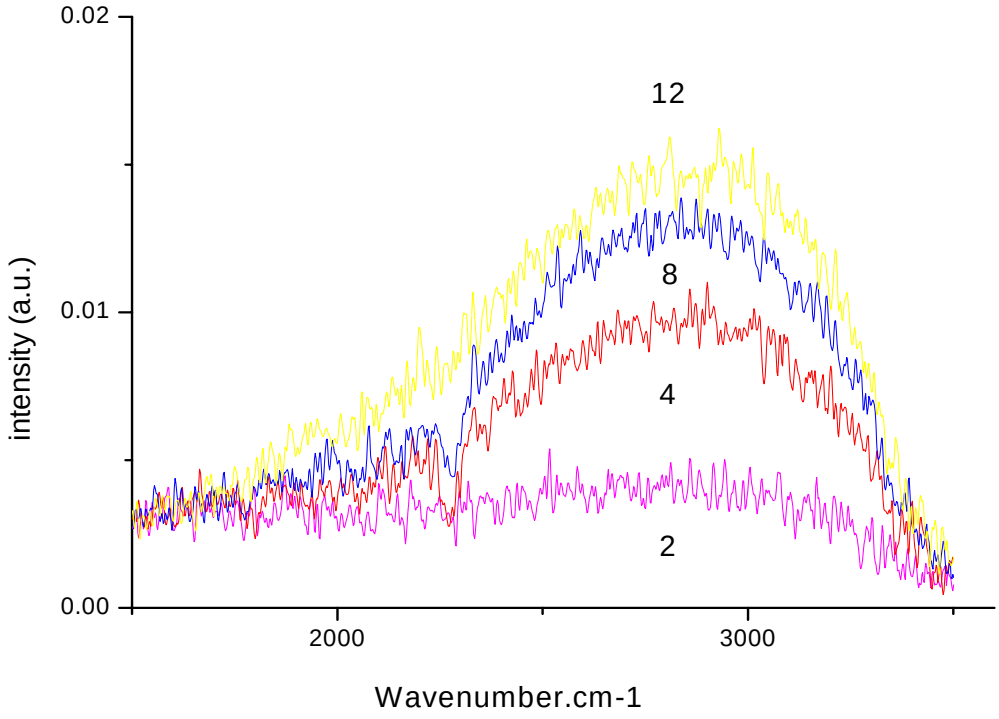

Figure (4.8); Neutron irradiated topaz for (2, 4, 8 and 12) hours, at OH band region

Fig (4-8) shows that the intensity of OH band increases by increasing the time of irradiation, which means more scattering of this band group and more collection of the cluster defects.

4.8.7. The effect of the thermal treatment on OH group

Raman spectroscopes were used by a number of authors to investigate the OH, SiO_4 and Al-related groups in topaz. In particular the modes associated with the SiO_4 groups are located around 935 cm^{-1}, while the lines in the region from 3625 to 3675 cm^{-1} are mainly due to different OH vibration modes [(103)]. While the bands at 3423 and 3486 cm^{-1} decrease, the bands at 3585, 3625 and 3650 cm^{-1} increase as the temperature of the thermal treatment increases; the other bands in the whole spectra, including the SiO_4 bands at 935 cm^{-1}, did not show any appreciable differences. The bands at 3625 and 3650 cm^{-1} are due to

crystallographically ‘‘normal’’ hydroxyl site while the others are normally attributed to OH associated with different defects. Thus the main effect of the thermal treatment is improving the ordering in the sample (enhancing of the intensities of the ‘‘normal’’ OH bands and weakness of those corresponding to the ‘‘defective’’ OH ions, as the temperature increases) [103]. The vibrational spectra suggest that the charge trapping centres are probably related to various OH related defect centres; the thermal treatments are mainly changing the distribution of the traps, rather than modifying the amount of recombination centres, enhancing the crystallographically ‘‘normal’’ hydroxyl ions [103]. Thus; the color centre which causing the color in topaz gem related directly to OH group movement. Also the thermal treatment changing the distribution of the traps, modify the amount of color centres leading to enhancing the color.

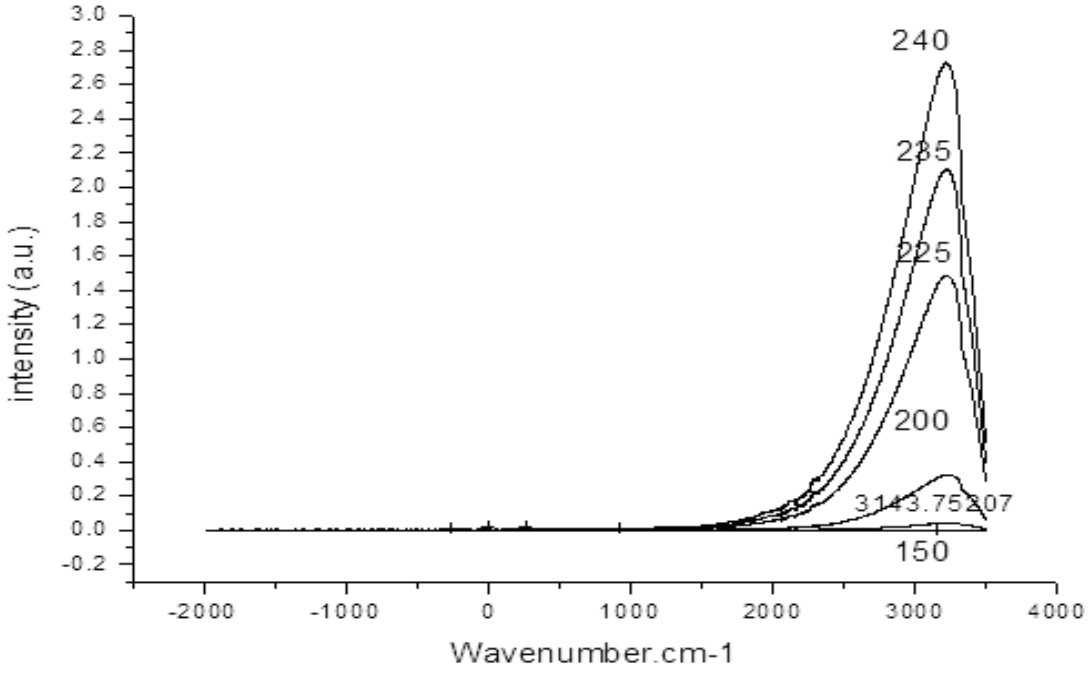

Figure (4.9); Topaz heating from 0 to 240 ^{0}C (for OH bands)

Inspection of Fig (4-9) shows that the intensity of OH group did not change by heating until 150 ^{0}C and it begins increasing continuously to 240 ^{0}C. The intensity of OH band increases by heating means that, more scattering of this band group and more collection of the cluster defects.

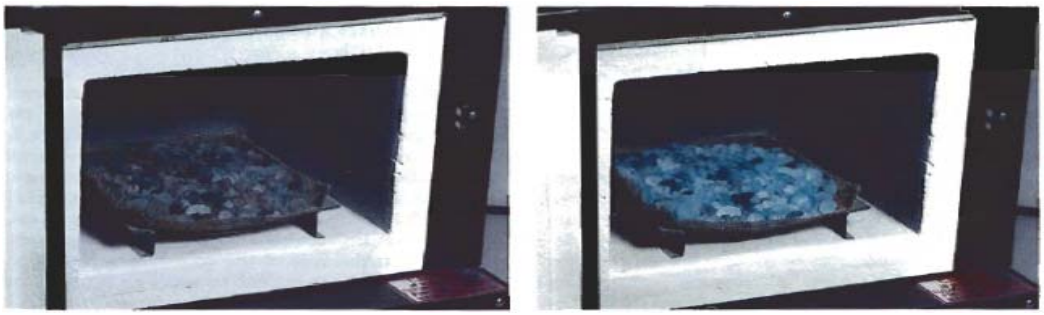

Fig (4.10) Topaz before (*left*) and after (*right*) heat treatment at 220 ^{0}C for (1.5) h.

Cooling topaz sample to -100 ^{0}C showed that the variation of OH group band intensity decreased by cooling, which means less scattering or decreases in cluster defects. The relation between temperatures in degree centigrade with intensity is a linear. On the other hand the study showed that the variation of intensity of OH group frequency with irradiation time increases continuously with increasing irradiation time up to 12 hours at the reactor. No investigation for more irradiation time but it is expected that up to certain time the intensity of OH group frequency could be constant whatever the irradiation time may increase. Also the study shows that, the Intensity of most bands in topaz irradiated by neutrons after γ irradiation are higher than the bands in topaz irradiated by γ. This is because, of the strong effect of neutrons which penetrates the atoms and interacts by impact with the OH groups leading to the deformations, while γ cause induced effects with the OH groups with less effect.

4.8.8. Topaz as irradiated by (γ, and n+ γ) results

Table (4.3); Bands and its Intensity in topaz as irradiated by (γ, and n+ γ)

Topaz irradiated by γ	Intensity	Topaz irradiated by neutrons after γ irradiation	Intensity
3938	27	3924	39
3874	113	3876	125
3768	75	3768	80
3760	62	3768	80
3702	33	3704	35
3598	12	3590	10
3524	57	3516	64
3436	12	3432	15
3224	44	3228	46
2996	19	2994	1
2550	751	2546	843
2462	673	2468	772
2368	193	2374	262
2084	1500	2124	181
2018	739	2088	1687
1880	174	1864	266
1660	1661	1664	1741
1588	227	1654	1844
1388	223	1388	444
928	233	934	705
852	481	838	976
664	247	668	765

Table (4-3) shows that the Intensity of most bands in topaz as irradiated by neutrons after γ irradiation are higher than the bands in topaz as irradiated by γ because of the strong effect of neutrons.

Table (4.4); some bands and its strength in topaz as irradiated by (γ, and n+ γ)

Band	*Strength degrees*
680	VW
925	W
1080	S
1175	W
1610	W
1820	VW
1880	VS
2100	W
2240	S
2320	VS
2590	W
2680	S
2780	S
3440	M
3700	S

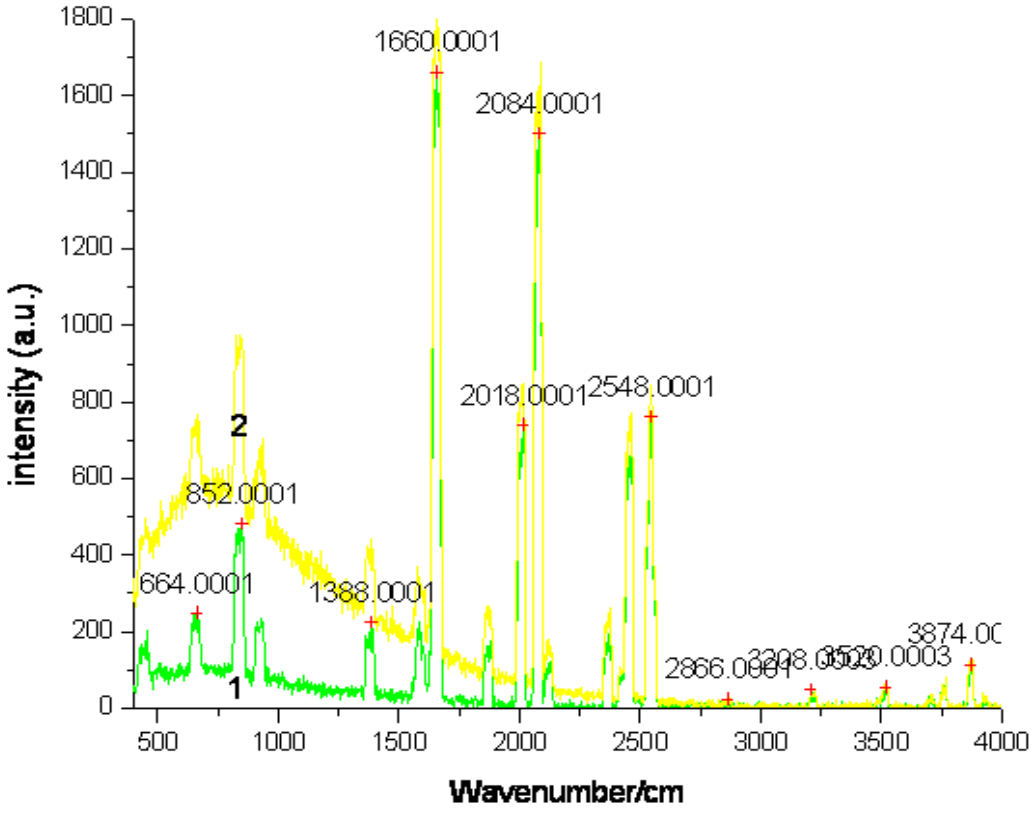

Figure (4.11); Topaz gamma irradiation (1) in green color and topaz (gamma + neutron) (2) irradiation in yellow color

Gamma radiation induced defects in the samples. These samples were investigated using FTTR Raman spectroscopy. The most intense Raman peaks were identified. The difference between these peaks of gamma irradiation and at states of raw and neutron irradiation at the same range were studied.

Table (4.5); Bands and its Intensity in Topaz

[Raw, irradiated (γ, n and n+ γ)]

(1) Topaz raw material,(2) Intensity, (3) Topaz irradiated by gamma, (4) Topaz irradiated by neutrons and (5) Topaz sample irradiated by neutrons after gamma irradiation and N means not measured.

R[(1)]	I[(2)]	γ[(3)]	I[(2)]	n[(4)]	I[(2)]	(γ+n) [(5)]	I[(2)]
268.368 vs	0.22071	N	N	268.37	0.21055	N	N
286.733 s	0.11252	N	N	285.72	0.19229	N	N
317.59 w	0.0117	N	N	316.58	0.0134	N	N
332.921 m	0.05478	N	N	332.01	0.01626	N	N
359.368 w	0.01713	N	N	372.51	0.00935	N	N
403.731 w	0.03442	404	0.0060	403.36	0.02885	404	0.0339
457.593 w	0.02612	454	0.0203	457.36	0.01587	458	0.0473
495.930 w	0.00554	494	0.0104	494.00	0.01191	496	0.0467
520.636 vw	0.01617	520	0.0103	521.00	0.01133	522	0.0515
548.668 w	0.01262	540	0.0110	549.93	0.01165	548	0.0498
561.2 w	0.01274	564	0.0103	561.49	0.03385	562	0.0524
620	0.00222	618	0.0109	619.35	0.00633	620	0.0582
643.3 vw	0.00498	664	0.0247	644.42	0.01155	668	0.0765
800	0.00274	802	0.0114	808.35	0.00565	806	0.0595
854. 98 m	0.01791	852	0.0481	856.56	0.00822	838	0.0976
925.76 vs	0.1224	928	0.0233	935.63	0.11968	934	0.0705
984.25 w	0.02371	984	0.0090	983.84	0.01917	988	0.0501
1008 w	0.01979	1004	0.0090	1005.05	0.00863	1010	0.0465
1079	0.00302	1078	0.0076	1089.91	0.00555	1084	0.0430
1164 w	0.00447	1172	0.0061	1163	0.00659	1178	0.0410
N	N	1388	0.0223	1388.82	0.00439	1388	0.0444
N	N	1588	0.0227	1587.46	0.00477	1654	0.1844
N	N	1660	0.1661	1660.74	0.00466	1664	0.1741
N	N	1880	0.0174	1863.23	0.00564	1864	0.0266
N	N	2018	0.0739	2021.37	0.00477	2088	0.1687
N	N	2084	0.1500	2081.15	0.00559	2124	0.0181
N	N	2368	0.0193	2374.28	0.00756	2374	0.0262
N	N	2462	0.0673	2474.56	0.00825	2468	0.0772
N	N	2550	0.0751	2545.92	0.00872	2546	0.0843

N	N	2996	0.0019	2995.26	0.00834	2994	0.0001
N	N	3224	0.0044	3224.75	0.00669	3228	0.0046
3143	0.00397	3150	0.0001	3155	0.0045	3150	0.0015
3423	0.00070	3436	0.0012	3423.38	0.00178	3432	0.0015
3486	0.00144			3488.95	0.00153	3486	0.0024
N	N	3524	0.0057	N	N	3516	0.0064
3585	N	3598	0.0012	N	N	3590	0.0010
3625	N	3628	0.0005	N	N	3622	0.0008
3650	N	3652	0.0007	N	N	3660	0.0007
3675	N			N	N	3682	0.0014
N	N	3702	0.0033	N	N	3704	0.0035
N	N	3768	0.0075	N	N	3768	0.0080
N	N	3874	0.0113	N	N	3876	0.0125
N	N	3938	0.0027	N	N	3924	0.0039

Raman spectroscopy was used to investigate the (OH, SiO_4 and Al) related groups in topaz. The modes associated with the SiO_4 groups are located around 935 cm^{-1} and the bands at 646, 847, 983 and 1163 cm^{-1} are related to the same group while the lines in the region from 3625 to 3675 cm^{-1} are mainly due to different OH- vibration modes. The two OH- stretching bands around 3640 and 3650 cm^{-1} appeared in other samples from different locations. It was noted that the bands at 3423 and 3486 cm^{-1} decrease while the bands at 3585, 3625 and 3650 cm^{-1} increase as the temperature of the thermal treatment increases. The other bands in the whole spectra including the SiO_4 band did not show any differences. The bands at 3625 and 3650 cm^{-1} are due to crystallographically normal hydroxyl site while the other are normally attributed to OH associated with different defects.

When the samples are irradiated, OH centres and Al^{3+} substituting the Si^{4+} centres act as charge trapping centres. The number of OH trapping centres could be changed by thermal treatment and irradiation.

4.9. Spectrophotometer:

The optical absorption spectra of raw and irradiated samples were carried out with "UV/VIS8500" spectrophotometer and measured in the spectral range from 190-1100 nm at room temperature.

The excitation device is equipped with a tungsten halogen/deuterium lamp. The emission is collected in a collimator, with resolution of 0.1 nm in the visible range, equipped with a photomultiplier. The excitation and emission slit width were dual-beam, single monochromatic grating system 1200 lines/mm.

The absorption of light is investigated by optical spectrophotometer at different irradiation and heating conditions. From the analysis, it is shown that the blue color induced by neutrons is independent of the origin of topaz. Its concentration and the blue color are increased nearly linearly as a function of the dose.

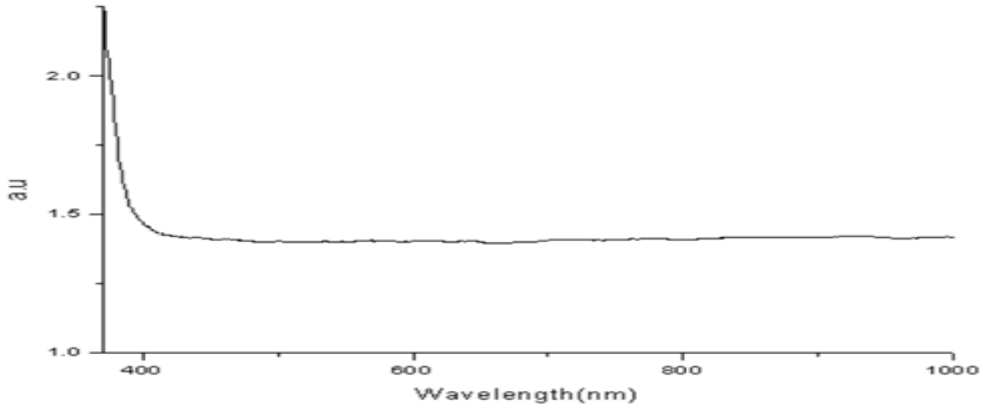

Fig (4.12); Optical absorption spectra of topaz at room temperature

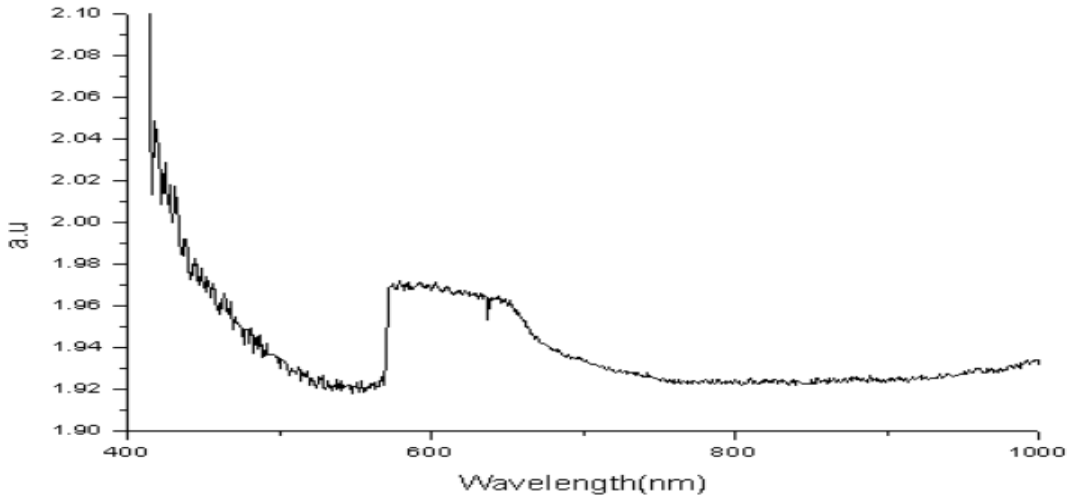

Figure (4.13) Optical absorption spectra of gamma irradiated topaz

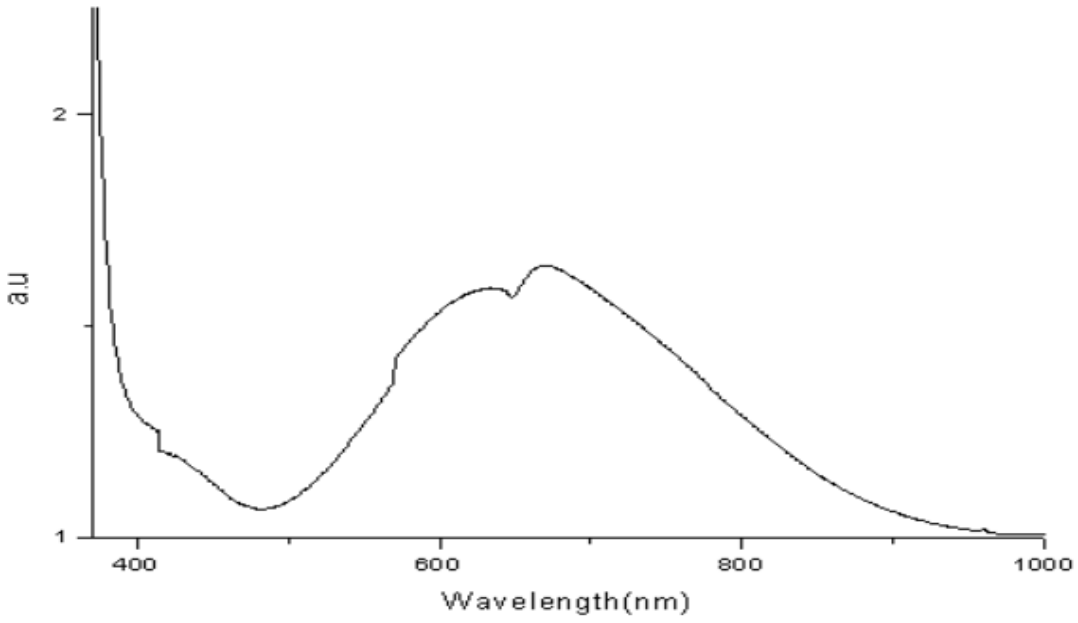

Figure (4.14) Optical absorption spectrum of neutron irradiated topaz for 12 hours

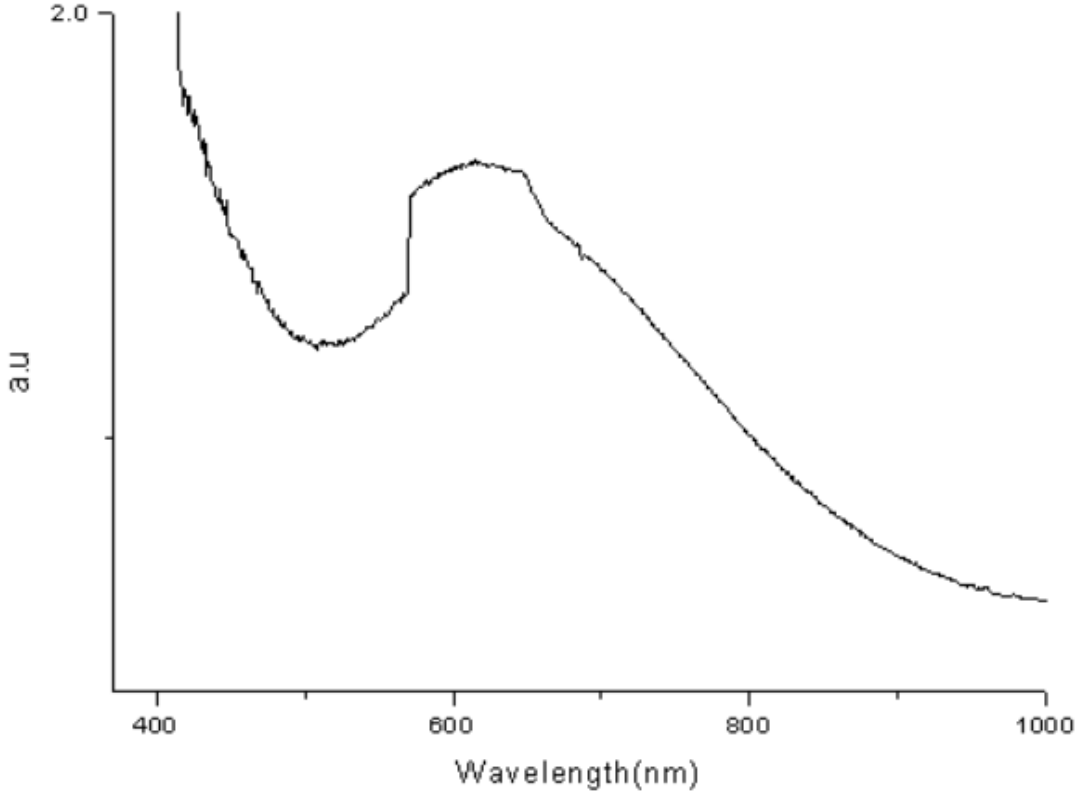

Figure (4.15) Optical absorption spectra of neutron irradiated topaz for 12 hours after heating at 200 C^0 for 2 hours

Natural topaz can be given a blue color in a stable way by different treatment processes as a result of the production of color centres that involve non-radiative transitions.

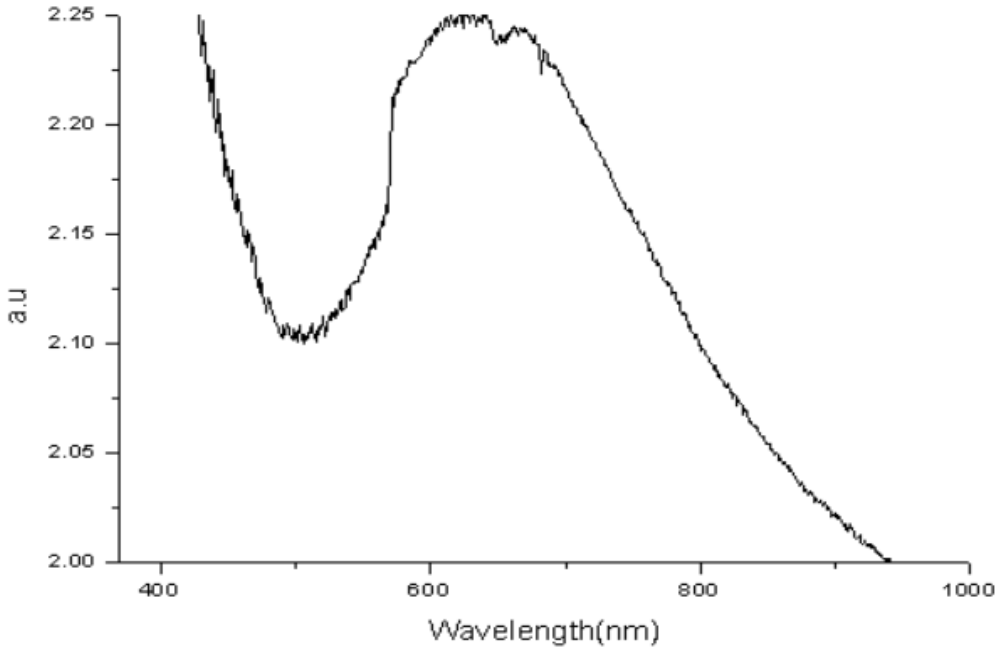

Figure (4.16) Optical absorption spectra of neutron irradiated topaz for 12 hours after heating at 200 C^0 for 4 hours

Topaz is predominantly crystallized in perfect prismatic crystals with the chemical composition $Al_2Sio_4(F,OH)_2$, with a rhombohedral structure. In its structure, all the hydrogen sites are symmetrically equivalent. The structure of the topaz is formed by monolayers of oxygen anions alternating with monolayers of F_2O in ABAC closed packaging sequence along the Y direction. One third of the octahedral sites are occupied by aluminum ions and 1/12 of the tetrahedral sites are filled with si^{4+}. The oxygen atoms are coordinated by one si^{4+} and two Al^{3+}, and the fluorine by two Al^{3+} ions [106].

The optical absorption spectra of samples were measured in the spectral range from 350 up to 800nm at room temperature. As can be noticed from the figures, the band centered above 600 nm increases its intensity with the dose. This band together with the absorption tail in the UV region is responsible for the blue color of the topaz leaving an absorption valley at about 480 nm. However, increasing the irradiation to the doubled time increases the absorption band intensity by only 10%. All the samples from different regions in Brazil

show the same optical absorption spectra, and the same trend on neutron irradiation, i.e., the absolute absorption is the same for all investigated samples. It means that the produced colors and color centres do not depend on the origin of samples.

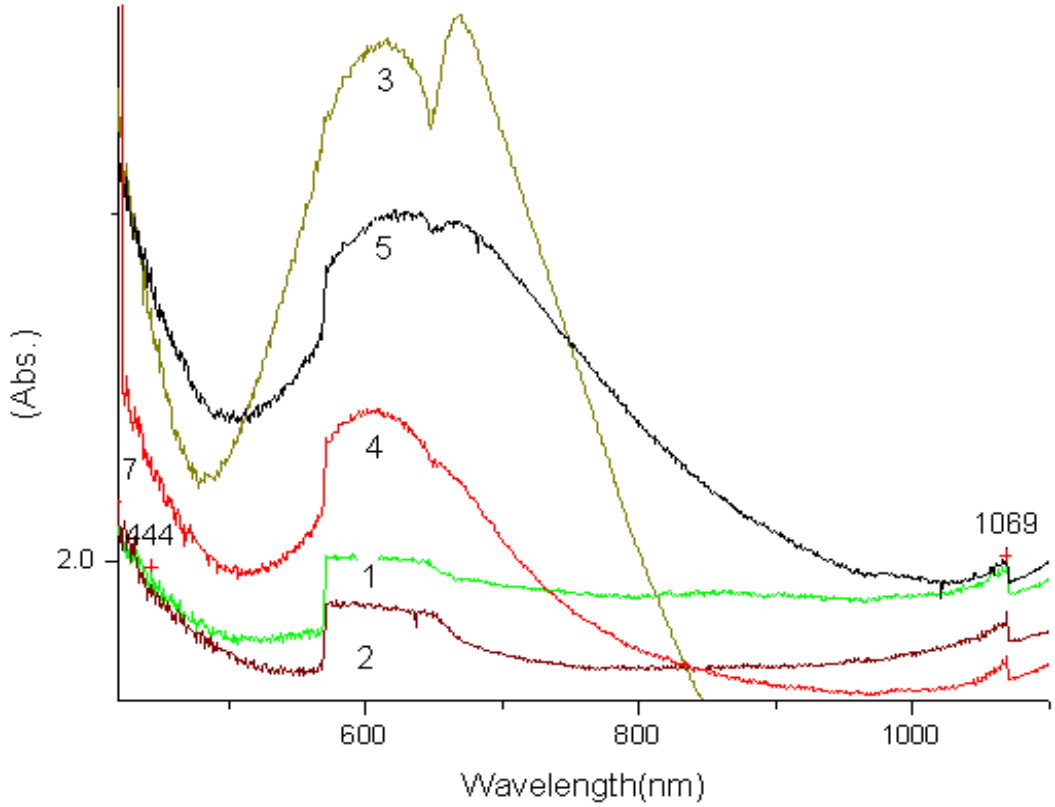

Figure (4.17); Topaz [1.Raw, 2. irradiated by γ, 3. irradiated by neutrons, 4. irradiated by neutrons after γ irradiation and 5. Heated gamma irradiated for 4 hours at $200C^0$]

The treatment of topaz stone with γ rays increases the overall optical absorption giving the crystals a brown color. Applying thermal treatment at range 200-250 0C, the brown component of optical absorption spectrum (~ 450-500 nm) is removed leaving a weak band around 650 nm, which gives the crystal in blue color. The apparent difference is thus the brownish color was more distinct in the neutron-irradiated and annealed stones. The treatment of

topaz with γ rays for months, continuously gives the blue color directly without heat treatment, but lighter than the color resultant from the reactor irradiation. It takes very long time but very low residual radioactivity, which goes to vanish quickly, through days. The apparent difference is thus the brownish which was more distinct in the neutron-irradiated and annealed stones.

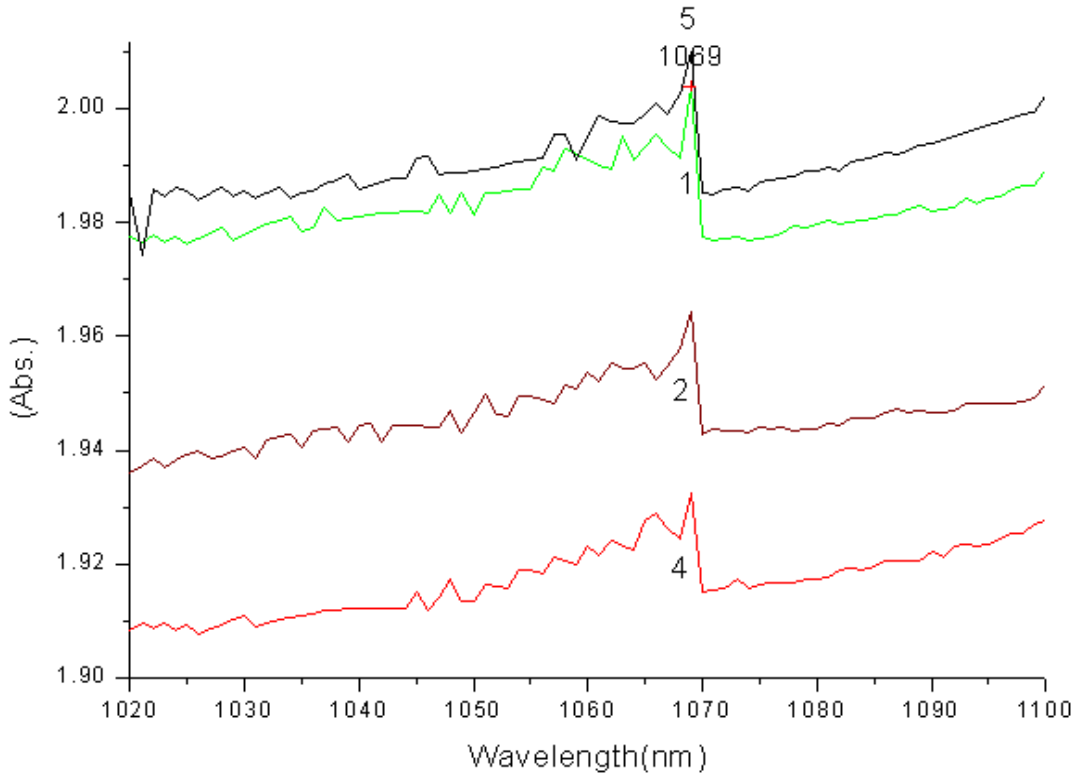

Figure (4.18); Topaz raw and treated (3 cases)

At the figure (4-20) "1" represents raw topaz, "2" is topaz irradiated by γ, and "4" is topaz irradiated by neutrons after γ irradiation and 5 represents heated gamma irradiated topaz for "4" hours at 200 C^0. It is notable that no beak at this value for topaz irradiated by neutrons; because it is vanish at 8440 approximately.

In the irradiated topaz there is a difference in the thermoluminescence; as the temperature is raised, there is an emission of light that is not

present in the natural blue topaz as reported [Petrov et al.(1977). Rossman G. (1981)] [5, 107].

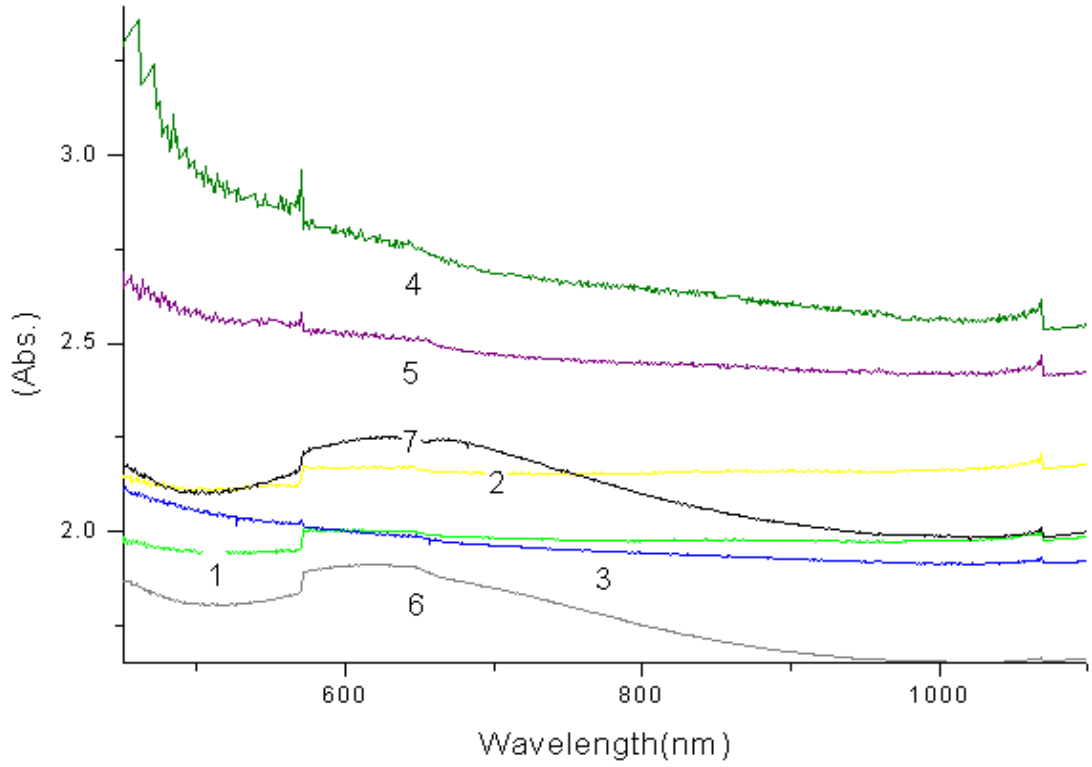

Figure (4.19); Topaz raw and treated (6 cases)

The intense absorption from the UV to about 480 nm is developed by irradiation and even increases in intensity after annealing. At figure (4-21) "1" represents raw topaz, "2" is raw topaz heated 1 hour at 200 C^0, "3" is raw topaz heated 2 hours at 200 C^0, "4" is topaz irradiated by γ and heated 2 hours at 200 C^0, "5" is topaz irradiated by γ and heated 4 hours at 200 C^0, "6" represents topaz irradiated by neutrons and heated 2 hours at 200 C^0 and "7" shows topaz irradiated by neutrons and heated 4 hours at 200 C^0. This figure shows that the absorption of the red color peak becomes wider and more stable after treatment with irradiation, and in addition to heat. This behavior is considered as an improvement in the topaz characterization.

4.10. Positron annihilation:

Irradiation-induced defects in natural topazes were investigated using positron annihilation spectroscopy. The present measurements are used to study the behavior of defect concentration in topaz in two states; pure and irradiated topaz by gamma and neutrons. It has been shown that positrons can become trapped in imperfect locations in topaz samples and their mean lifetime can be influenced by changes in the concentration of such defects. No changes have been observed in the mean lifetime values after the saturation of defect concentration. The mean lifetime and trapping rates were studied for three samples.

Since 1960, evidence has been collected that positrons can become trapped in imperfections in solids. This conclusion was based upon observation that the characteristics of the annihilation process could be influenced by changes in the concentration of defects in the solid in which the annihilations occurred. For example, the spectrum curves for the mean lifetime of positrons change as a function of the sample deformation or annealed temperature [42, 108]. Advantages of these methods in obtaining both qualitative and quantitative data on behavior are pointed out. To explain these results, several groups [109:110] had proposed a phenomenological trapping model which actually had been introduced earlier by Brandit [111]. The main assumption of the simple trapping model is that positrons annihilate in a solid from a free or trapped state. Escape from traps is neglected, where the positrons are radiated during the beta decay of certain artificially produced radioactive isotopes such as ^{22}Na. A complete isolation positron is a stable particle, but in a solid it has a very short lifetime, less than one nanosecond, where it undergoes an annihilation reaction with an electron. In the reaction the two particles (electron-positron) disappear and their total rest mass energy (1.022 MeV) appears as the emission of two gamma rays, each with an energy of almost 0.511 MeV. These rays emerge from the point of annihilation in exactly two opposite directions. The present work reports the

results of the positron annihilation investigation on 3 samples of topaz; pure, irradiated by γ and neutron irradiated samples.

^{22}Na radioactive isotope is used in the present work as a positron (β+) source. It was evaporated from an aqueous solution of free carrier ^{22}NaCl with 1 mCi activity, and deposited on a thin kapton foil of 7.5 μm thickness. ^{22}Na decays with the emission of a β+ with 511 KeV followed by a 1274 KeV γ-ray.

A sandwich configuration has been used for samples measurements. The present studies were performed using sets of couple of two identical samples of the material under investigation.

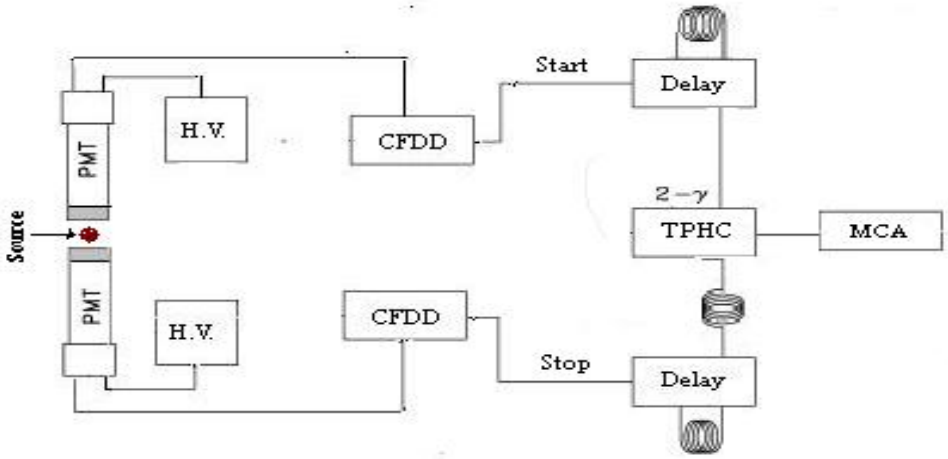

Figure (4.20); Schematic diagram for the detection of the 2-γ positron annihilation.

The schematic diagram for the Fast coincidence system which was used in the measurements is shown in figure (4.20), with time resolution of 290 ps [112, 113]. The equipment consists of a pair of scintillators and photomultipliers (PMTs) rigidly mounted. Positive high voltage power supply provides power to the PMTs via voltage divider which allows one to vary the potential applied to the two tubes. The 1274 keV γ-ray was taken as a start signal for the time to pulse height converter (TPHC), while one of the 511 keV annihilation quanta, resulting from the 2γ-rays annihilation in the material, was chosen as the stop

signal. The signal from the TPHC was fed to the multi-channel analyzer (MCA) for storage. The 1274 keV signal indicates the birth of the positron and the 511 keV indicates its death. Thus, the time interval between the above 2γ-rays was the lifetime of the positron or Ps in the material. The thickness of the samples is adequate to absorb the emitted positrons. For most of measurements, account period at least 3 hrs were required to obtain sufficient statistics. The technique of positron annihilation (lifetime technique) is described in more details elsewhere [42].

The lifetime spectra were analyzed as 2 lifetime components (τ_1, and τ_2), with corresponding intensities I1and I2 with the help of computer program PATFIT-88 [114]. The positron source is sandwiched between two identical samples. The source-sample configuration was then wrapped in a thin aluminum foil. Each sample was measured for 10800 seconds during which about $3.17x10^5$ coincidence events were accumulated.

4.10.1. Experimental Procedures

At Positron Annihilation Spectroscopy (PAS) The Fast-Fast coincidence technique, which is described elsewhere [42, 76], and the positron lifetime measurement with a half maximum (FWHM) time resolution of 290 ps was used. The 1274 keV γ-rays were taken as the start signal for TPHC or the "time to pulse height converter" (TPHC), while one of the 511 keV annihilation quanta, resulting from the two photons annihilation in the material, was chosen as the stop signal. The signal from the TPHC or the "time to amplitude converter" (TAG) was fed to the multi-channels analyzer (MCA) for storage. The 1274 keV signal indicates the birth of the positron and the 511 keV annihilation energy indicates the positron death. Thus, the time interval between the above two γ-rays is the lifetime of the positron in the material.

The resolution function of the system could be measured by using the coincidence as given by the ^{60}Co source. Because there is no time difference (simultaneous) between the two γ-rays emitted from the decay of ^{60}Co, it is preferable for measuring the resolution of the system. Such a resolution function describes the quality of the lifetime measuring system. This is normally described by the full width at half maximum (FWHM) of the prompt curve which is created by the Coincidence between the start and stop signals at a fixed tie delay. The prompt curve can be seen as a Gaussian distribution.

The positron lifetime was measured for pure and irradiated topaz. The dimensions of topaz samples were: (1 x 1 x 0.2) and the thickness was around 2.2 mm. The positron source of 1mCi free-carrier $^{22}NaCl$ was evaporated from an aqueous solution of sodium chloride and deposited on a thin Kapton foil of 7.5 μm thickness. The positron source is sandwiched between two identical samples.

Raw and irradiated samples were measured for the same time; 010800 seconds. The data for the lifetime spectra were analyzed by using the PATFIT88 computer program [114]. The value of the source contribution (Kapton foil) of lifetime and its intensity was subtracted during the analysis. The block diagram of the system used in measuring lifetime is described elsewhere [76].

4.10.2. Results and Discussion

The results of the positron annihilation experiment on topaz samples, which contain defects that trap positrons, were analyzed. The main radiation –induced defects in topaz were previously observed and identified by Petrov [112]. The dislocation density (ρ) is generally expressed as length per unit volume, (ρ cm^{-2}) [115]. The defect density is related to dislocation density.

Table (4.6); Lifetime of the positron in topaz raw, irradiated by γ and irradiated by neutrons

Sample name	Lifetime (PS)	Intensity	SD for lifetime (PS)
Topaz raw	188.1	86.02	± 9
Topaz irradiated by γ irradiation	189	86.55	± 8.5
Topaz irradiated by neutrons	198.6	87.05	± 7

Because of the vacancy clustering the signal increases in positron lifetime and table (4-6) shows the variation in the lifetime of the positron in topaz as raw, irradiated by γ and irradiated by neutrons. The data means that the deformation in topaz which irradiated by neutrons has the largest effect leading to deformation and rearrangement and, consequently causing deepest and more stable color.

Gamma interaction with topaz sample is done only between γ rays and outer levels of electrons, thus; the result of this interaction is weak compared to the effect of neutrons interaction and the resultant color appears slightly and on topaz surface.

Positrons are injected into topaz sample body their lifetime strongly depends on whether they end up in a region with high electron density or in a void where electrons are scarce or absent. In the latter case the lifetime can be much longer because the probability to run into an electron is much lower. Irradiation causes remarkable changes in the annihilation characteristics depends on the irradiation type. Defects exist where atoms are missing or their density is locally reduced, so, the repulsion between the positron and ion cores is decreased. Also, the redistribution of electrons causes negative electrostatic potential at this type of

defect. Thus positrons see defects like vacancies, voids and dislocations as strongly attractive centers in the crystal.

By comparing the fraction of positrons that have a longer lifetime to those that annihilate quickly one can therefore gain insight in the voids or the defects of the structure. Then, defects are increase from raw topaz to topaz irradiated by gamma and finally goes to its maximum value after irradiated topaz by neutrons.

This deference is due to the cluster of vacancies. The study of defects in topaz of various colors still very littlie. This study aims to find relationship between the defects and color in irradiated gamma topaz crystals.

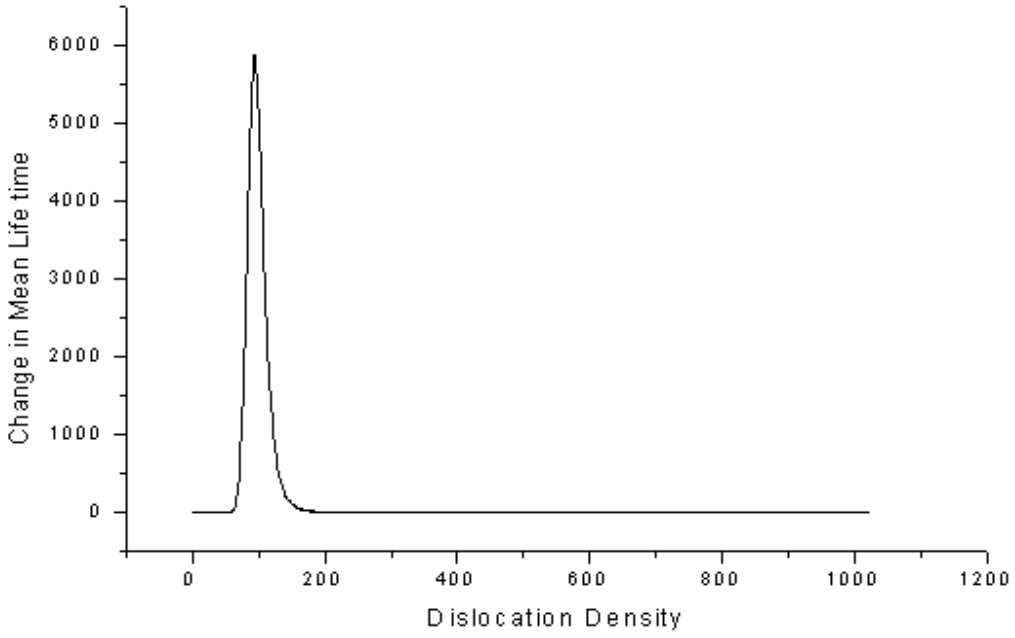

Figure (4.21); Change of mean lifetime as a function of dislocation density in pure topaz

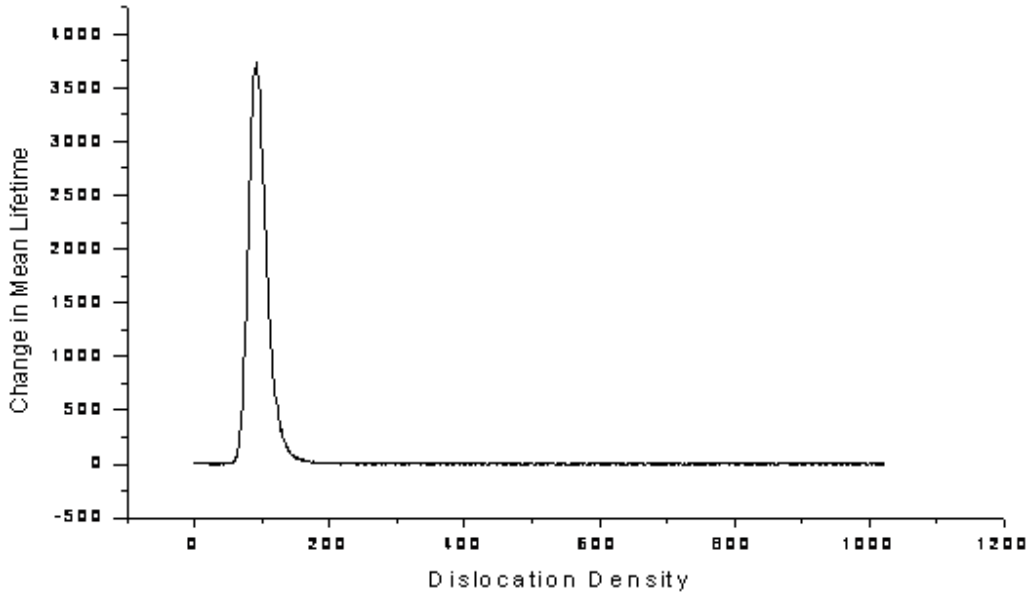

Figure (4.22); Change of Mean Lifetime as a function of dislocation density in Topaz irradiated by gamma

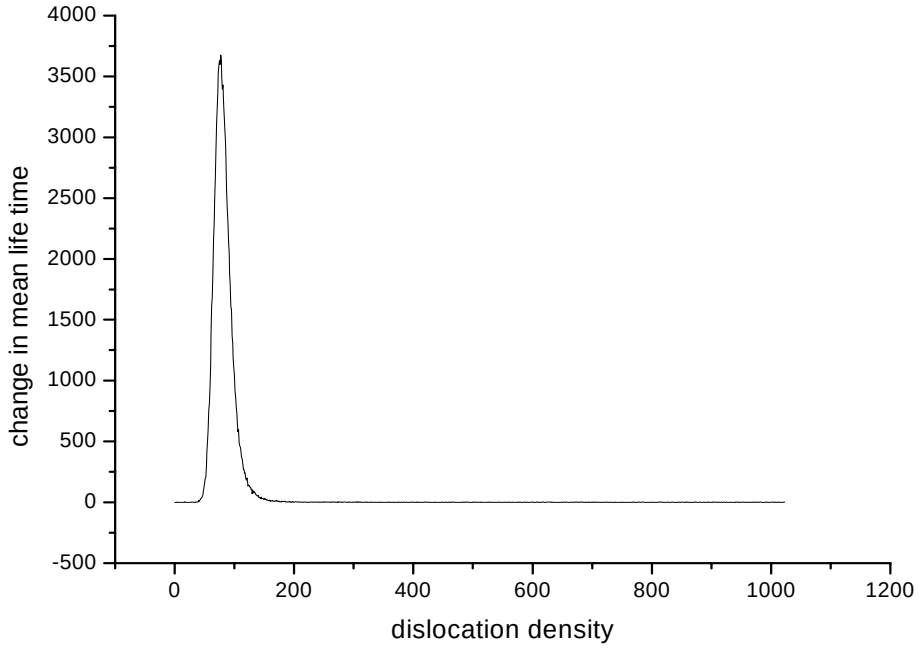

Figure (4.23); Change of Mean Lifetime as a function of dislocation density in Topaz irradiated by neutrons

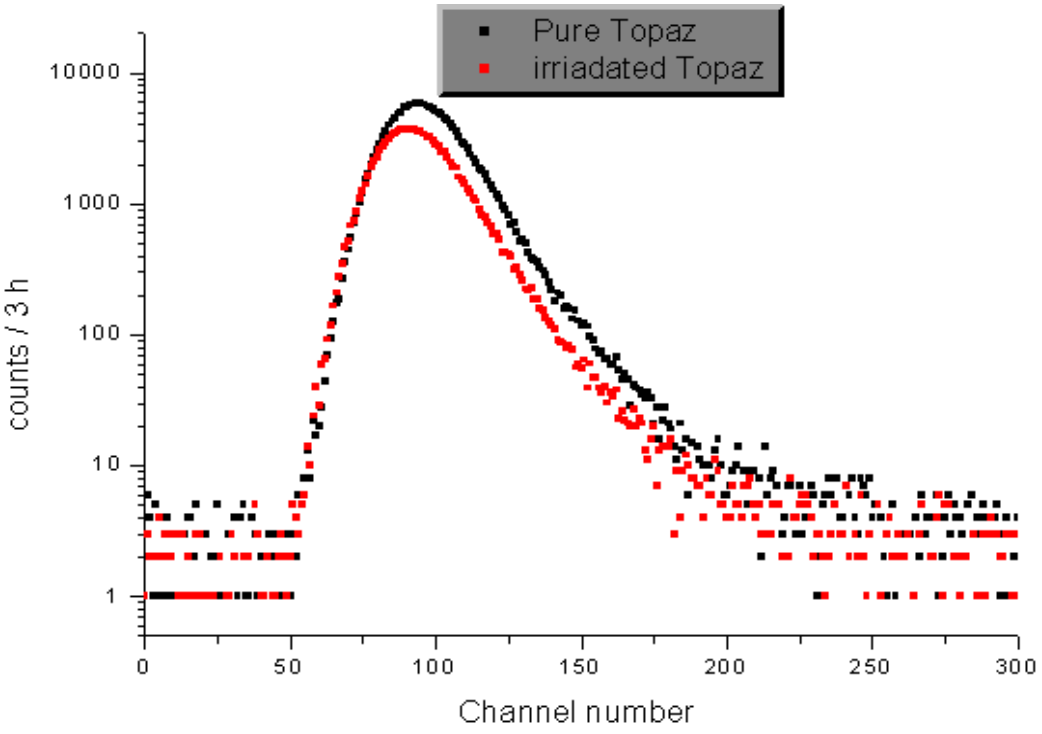

Figure (4.24); PAS curve of topaz (pure and irradiated by gamma)

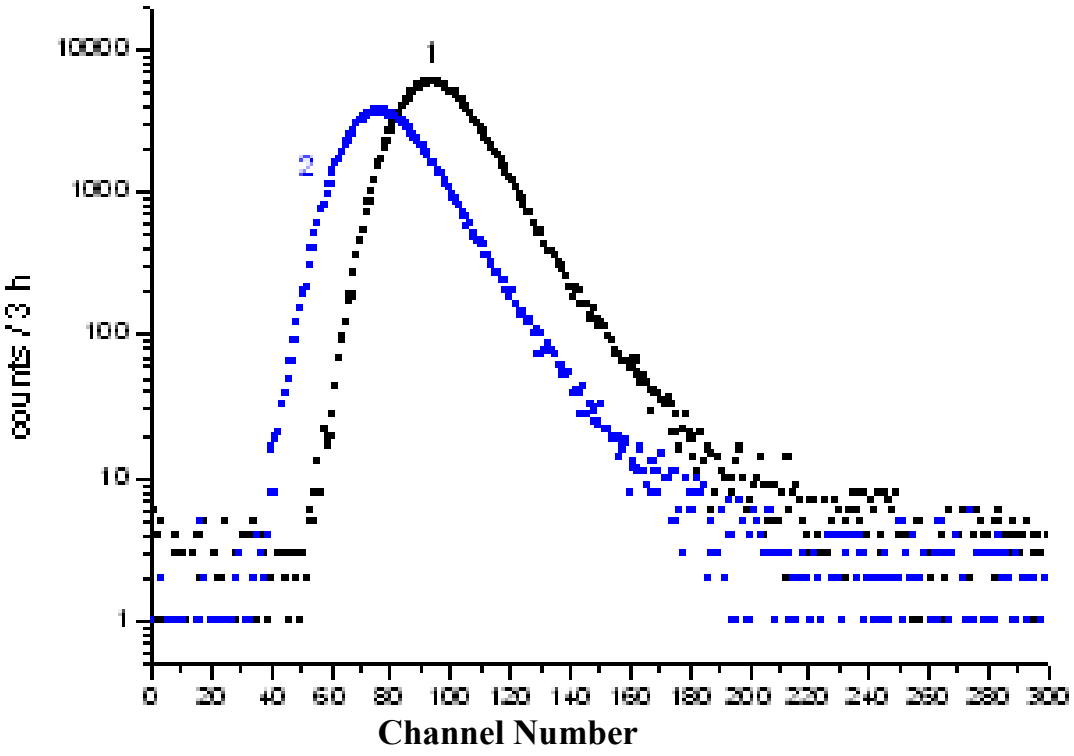

Figure (4.25); PAS curve of topaz [pure (1) and irradiated by neutrons (2)]

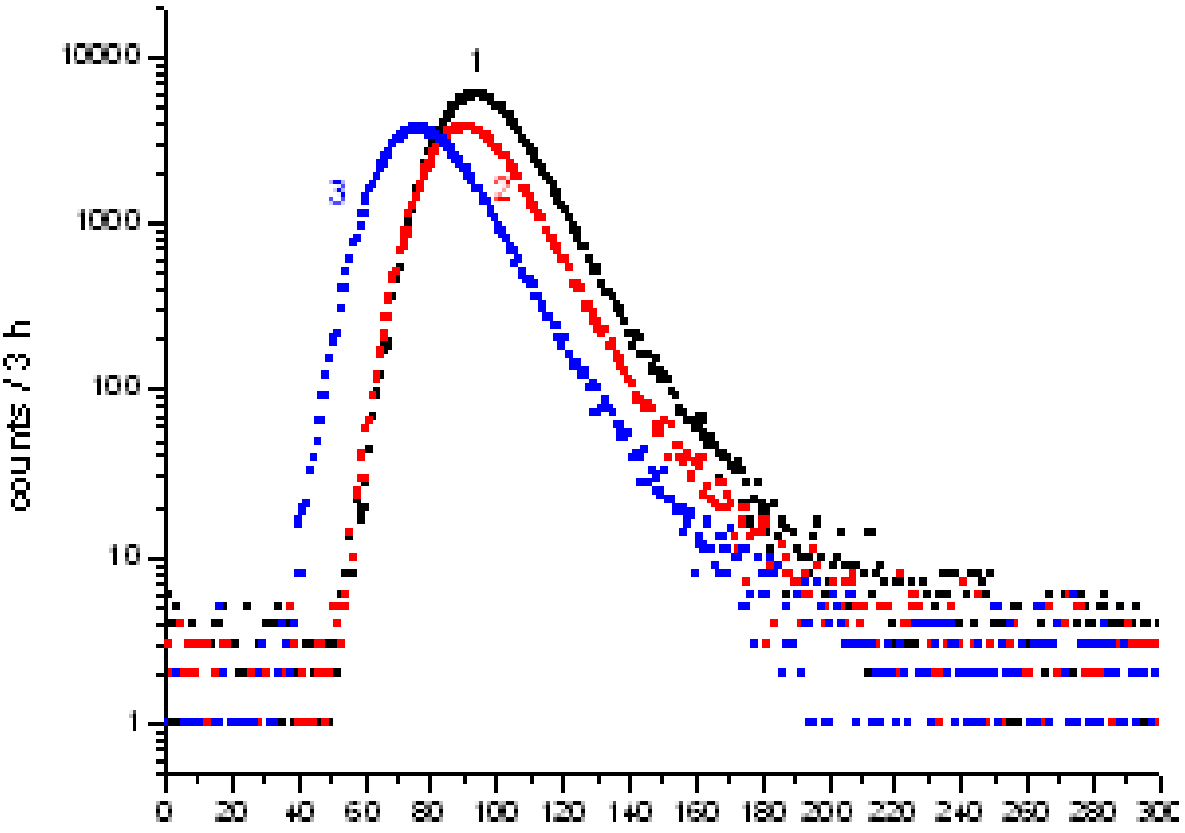

Figure (4.26); PAS curve of topaz [pure (1), irradiated by γ (2) and irradiated by neutrons (3)

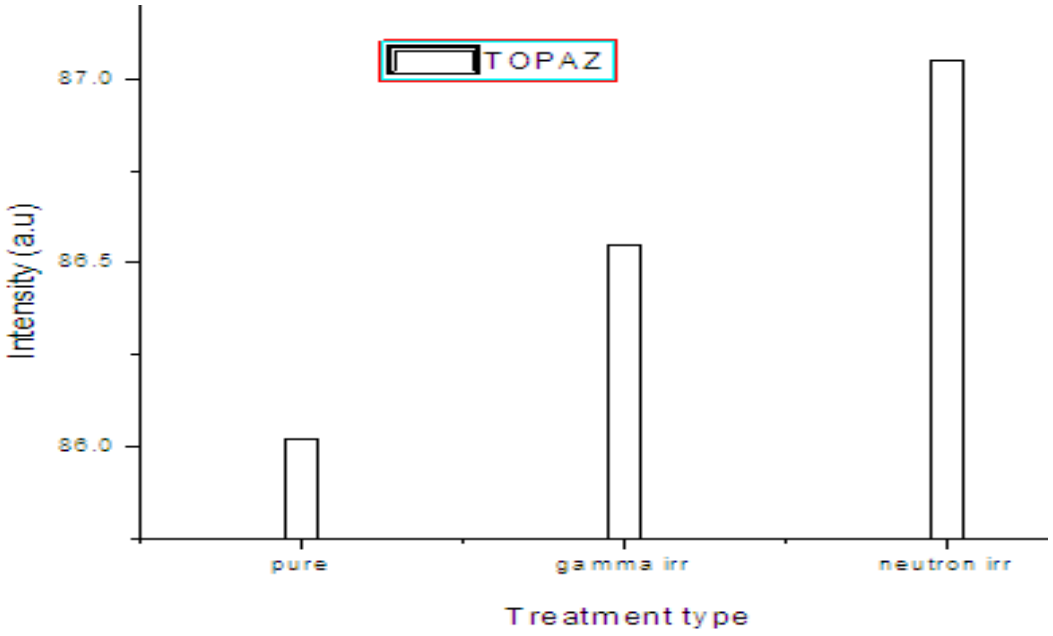

Figure (4.27); Change of intensity with treatment type

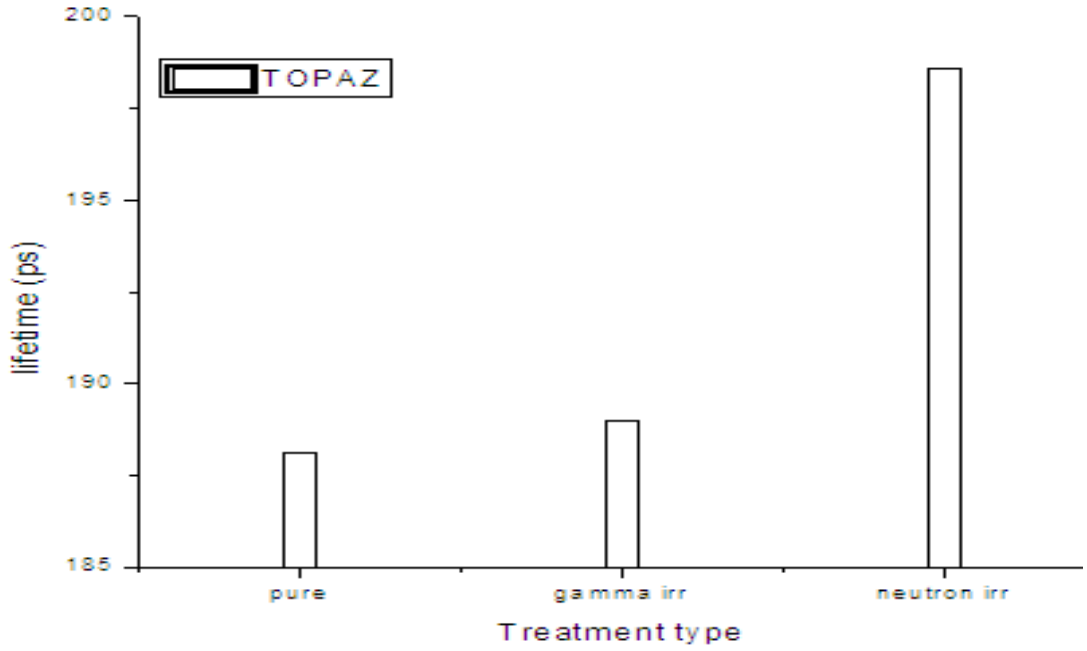

Figure (4.28); Change in lifetime with treatment type

It is possible to create more complex defects by irradiating topaz with long periods of irradiation times. The fact that the natural color is related to vacancies due to deformation of topaz while the vacancies are more evenly distributed by irradiation could explain why these types of blue color are differently distributed while they may be caused by a similar and very stable defect consisting of multiple vacancies (vacancy cluster).

4.11. Radiation Protection

4.11.1. Radiation Detection Equipment results

Irradiated gemstones are currently processed for color improvement by subjecting clear stones to neutron or high-energy electron irradiations, which leads to activation of trace elements in the stones. Assessment of the risk to consumers required the identification and quantification of the resultant radionuclides and the attendant exposure. Representative stones were irradiated and analyzed for gamma ray and beta particle emissions, using germanium spectrometer and Geiger-Muller counter. Based on these studies and other information derived from published literatures, dose and related risk estimates were made for typical user conditions. New criteria and methods for routine assays for acceptable release based on gross beta and gross photon emissions from the stones [(9)].

"Thermo FH 40 G" radiameter model "DB-033-961017 E" exposure rate measuring unit with FH 40 TG teleprobe was used for these measurements (table 4.7).

Table (4.7); portable telescopic probe survey meters measuring results

Sample name	**Exposure rate (at contact) (μSv/h)**	**Exposure rate (at 1 meter) in (μSv/h)**
Topaz irradiated by neutrons	0.3	0.02
Topaz irradiated by neutrons after γ irradiation	0.36	0.024
Topaz irradiated by neutrons twice	0.56	0.14

Portable contaminations monitor "CONTAMAT" FHT 111 M was used for these measurements (table 4.8).

Table (4.8); portable contamination survey meter measuring results

Sample name	**Surface contamination (Bq/cm^2)**
Topaz irradiated by neutrons	0.35
Topaz irradiated by neutrons after γ irradiation	0.57
Topaz irradiated by neutrons twice	0.73

The alpha content of raw samples was determined during long time of counting in the hours range, using a contamat. No alpha activity above background was detected. The CONTAMAT FHT 111 M was used for these measurements.

4.11.2. Cooling time calculations

Calculations were done using FORTRAN Program dependening on Newton-Raphson method.

The Newton Raphson algorithm is an iterative procedure that can be used to calculate maximum likelihood estimates (MLEs). The basic idea behind the algorithm is the following. First, construct a quadratic approximation to the function of interest around some initial parameter value. Next, adjust the parameter value to that which maximizes the quadratic approximation. This procedure is iterated until the parameter values stabilize [(77)].

In numerical analysis, the Newton-Raphson method is perhaps the best known method for finding successively better approximations to the zeroes (or roots) of a real-valued function. Newton's method can often converge remarkably quickly; especially if the iteration begins "sufficiently near" the

desired root. Just how near "sufficiently near" needs to be, and just how quickly "remarkably quickly" can be, depends on the problem.

Given a function $f(x)$ and its derivative $f'(x)$, we begin with a first guess x_0. A better approximation x1 is:

$$x_1 = x_0 - \frac{f(x_0)}{f'(x_0)}.$$

The program designed on "Microsoft developer studio, featuring Fortran Power Station" software.

The total activity of the stone is (A) which is the sum of activities of its components and the required is the time of cooling which verify the transportation limit of activity which is 74(Bq/g) [28] that represents the safe level transportation on air according to IATA regulations.

$$f(\chi) = \sum_{i=1}^{n} A_i e^{-\lambda_i x_i} \quad \rightarrow \quad (1)$$

$$f = 74 \text{ Bq / g} \quad \rightarrow \quad (2)$$

$$f'(\chi) = -\sum_{i=1}^{n} \lambda_i A_i e^{-\lambda i \chi i} \quad \rightarrow \quad (3)$$

From Newton Raphson algorithm A better approximation x_1 is:

$$\chi_1 = \chi_0 - \{[f(\chi_0)]/[f'(\chi_0)]\} \quad \rightarrow \quad (4)$$

So;

$$\chi_1 = \chi_0 - [A(\chi_0) / A'(\chi_0)] \quad \rightarrow$$

(5)

$$\chi_2 = \chi_1 - [A(\chi_1) / A'(\chi_1)] \quad \rightarrow \quad (6)$$

And so on till:

$$\chi_n = \chi_{n-1} - [A(\chi_{n-1}) / A'(\chi_{n-1})] \quad \rightarrow \quad (7)$$

Apply the condition that the error must be achieving the following;

$$x_n - \chi_{n-1} \leq \sum 10^{-4} \quad \rightarrow \quad (8)$$

Then the value of χ_n is obtained when A = 74 Bq / g

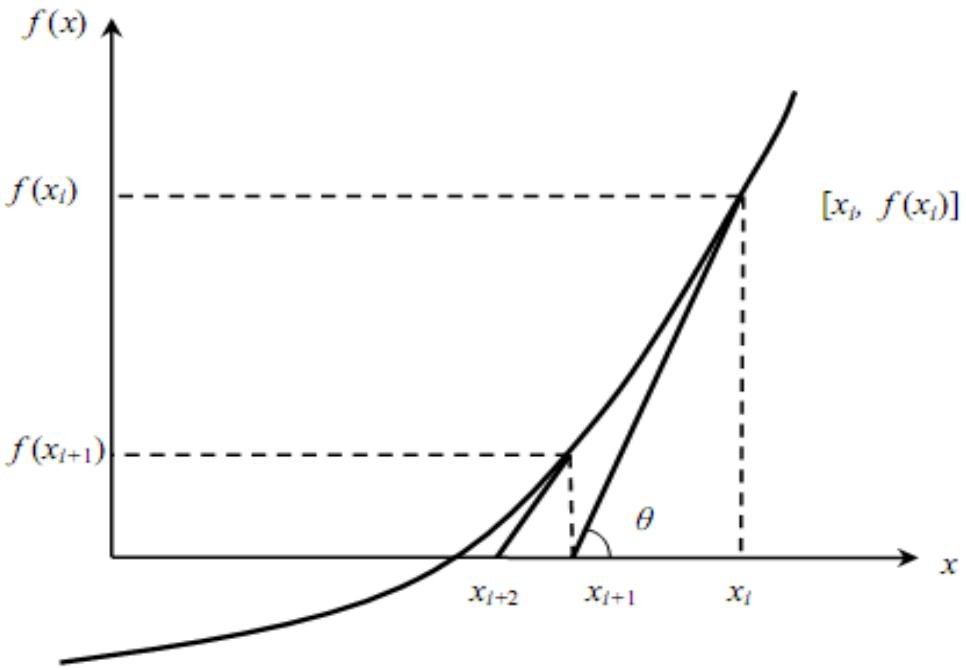

Figure (4.29); Newton Raphson algorithm better approximation

At figure (4-29) X-direction presents "t" and Y-direction presents "f = A (t) ". This figure illustrates one iteration of Newton's method (the function f is shown in curve x_{i+2} and the tangent line is represents $x_{i+1}=t_{n+1}$ and pointed line represents $x_i=t_n$). It is clear that t_{n+1} is a better approximation than t_n for the root x of the function f.

Table (4.9); calculated cooling time of some samples

Sample	**Time* (yr)**
Topaz irradiated by neutrons	1.386
Topaz irradiated by neutrons after γ irradiation	1.520
Topaz irradiated by neutrons twice	2.318

* Time (yr) required for activity to reach the safety level for transportation by air according to IATA regulations (74 Bq/g) [28].

* General decay any sample after (γ, neutron, γ +neutron and neutron twice) irradiation increase gradually.

Topaz as irradiated in a nuclear reactor contains many types and amounts of radionuclides as reported before [28]. It takes to be releasable, after radioactive decay 1.38 year according to table (4-9) while it takes a little more than two years after irradiation [66] at some cases.

Table (4.10); Isotopes in the sample of Topaz irradiated by neutrons

Isotope	A_e	Radiation emitted	Half-life (days)
Sc-46	652.24	β+ γ	83.79
Co-60	18.85	β + γ	1925.37885
Ta-182	18.85	β + γ	114.43
Fe-59	24.68	β + γ	44.503
Mn-54	11.73	γ	312.3

Where element mass (m_e) = 0.46484 g.

γ is the more effective from the radiation protection point of view because of its high ability for penetration while β could be protected easily. Between all isotopes in topaz Co-60 is highly considered because of its very long half life which increases the time of decay even if its concentration is low.

Table (4.11); Activities in the sample of Topaz irradiated by (n+ γ)

Isotope	A_e (Bq/g)
Sc-46	3338.58
Co-60	4.98
Ta-182	39.78
Fe-59	21.8
Mn-54	4.03

Where element mass (me) = 0.54778 g.

Usually the activity of Sc-46 at most times for most samples is the higher than the sum of all other activities. The ratios in activities differ according to the concentrations of these isotopes in the samples according to its origin.

Table (4.12); Specific activities of some isotopes in topaz as a function of time

	Time (days)	**Specific activity (Bq/g)**				
Isotope $T_{1/2}$ **(days)**		**Sc-46** 83.81	**Cs-134** 754.1	**Ta-182** 114.43	**Fe-59** 44.51	**Mn-54** 312.1
	20	70.98	8.21	251.06	0.44	7.56
	40	70.11	8.12	242.3	0.43	7.35
	60	68.6	8	196.2	0.42	6.85
	80	56.8	7.84	182.2	0.39	6.31
	100	54	6.11	135.55	0.36	5.71
	120	52.9	5.17	112.8	0.31	4.95
	140	51	4.88	95.3	0.22	3.83
	160	47	4.2	94	0.18	2.66
	180	22.89	3.63	86.44	0.04	2.23
	200	20.39	2.62	77.04	0	1.87
	220	19.79	2.44	75.51	0	1.09
	240	19.26	2.08	66.94	0	0.84
	260	18.07	1.84	65.84	0	0.66
	280	17.86	1.7	58.89	0	0.38
	300	17.34	0.945	56.89	0	0.35
	320	16.98	0.754	48.73	0	0.32
	340	16.46	0.633	45.98	0	0.28
	360	15.53	0.219	43	0	0.25
	380	14.46	0.18	41	0	0.21
	400	9.82	0.09	33.25	0	0.19

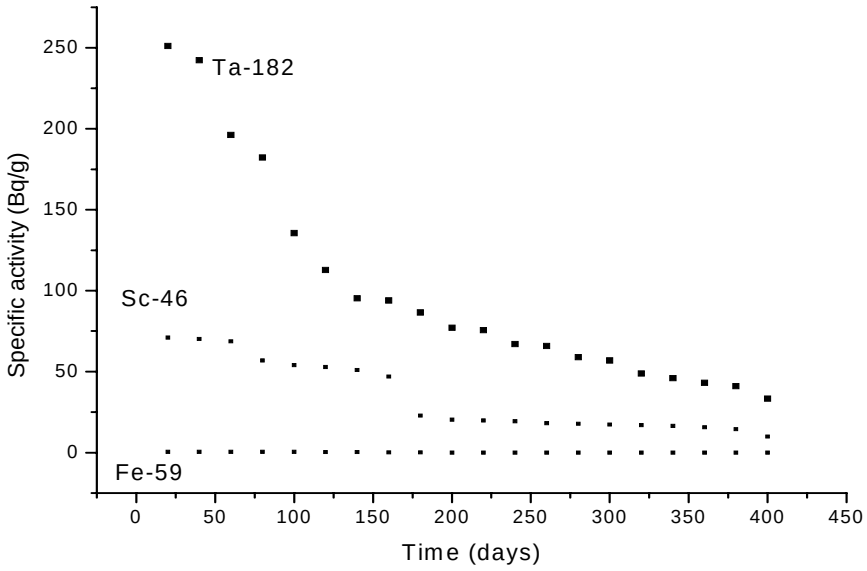

Figure (4.30); Decay of radioisotopes in topaz sample

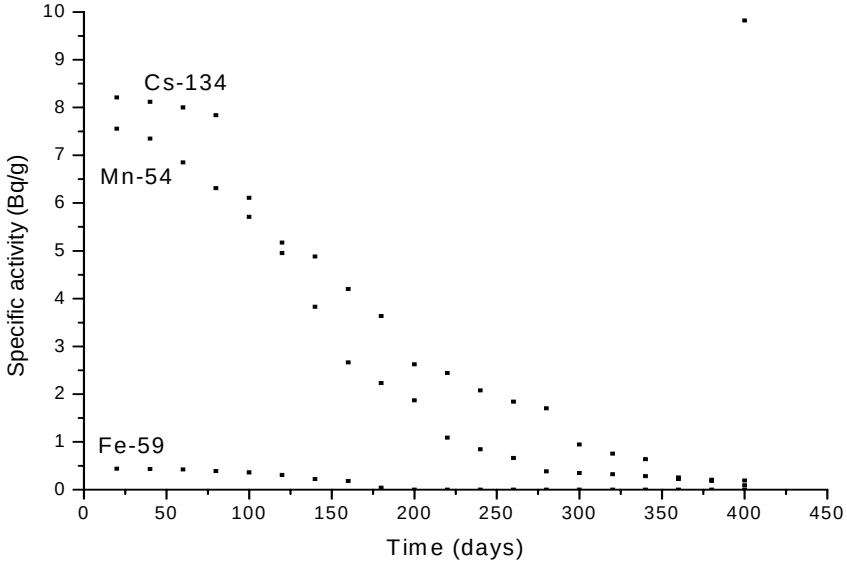

Figure (4.31); Decay of (Cs-134, Mn-54 and Fe-59) in topaz sample

Table (4.13); Decay dependent on type of treatment and calculation

Decay dependent on treatment type and calculation	Time (years)
Topaz irradiated by neutrons (Experimental)	1.12
Topaz irradiated by neutrons (Experimental considering all isotopes)	1.39
Topaz irradiated by neutrons (Theoretical)	1.42
Topaz irradiated by neutrons after gamma irradiation (Theoretical)	1.52
Topaz irradiated by neutrons twice (Theoretical)	2.31

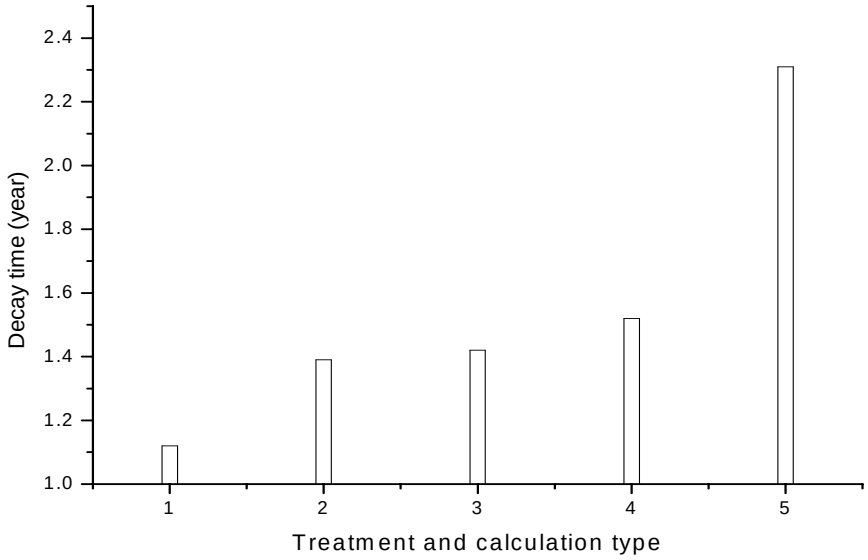

Figure (4.32); Decay dependent on type of treatment and calculation

4.11.3. Concentrations calculations of isotopes in samples

One of the objectives of this study is to calculate the concentrations of isotopes in neutron irradiated topaz. This is important to identify the specific nuclides responsible for the radiations. It helps to calculate the dose to people attributable to the induced activity.

Table (4.14); Survey of concentrations calculations of topaz irradiated by neutrons

Isotope	A_e	m_e	A_s	m_s	Specifc activity (A_e)	Specific activity (A_s)	C_s (ppm)	C_e (ppm)
Sc-46	652.24	0.46	64.84	0.14	1403.15	4.48E+02	7.24E+00	2.27E+01
Co-60	18.85	0.46	13.78	0.14	40.55	9.51E+01	2.00E-02	8.53E-03
Ta-182	18.85	0.46	31.09	0.14	226.70	2.15E+02	4.40E-01	4.65E-01
Fe-59	24.68	0.46	1.85	0.14	53.09	1.28E+01	5.01E+03	2.08E+04
Mn-54	11.73	0.46	2.05	0.14	25.23	1.42E+01	8.00E-02	1.43E-01

***(Cs)** in (ppm) is (Certified concentration of element in reference material) and (Ce) is Concentration of interested element.

Table (4.15); Comparison between concentrations calculations of topaz irradiated by neutrons by using Comparator and Relative methods

Isotope	Concentration (ppm)	
	Comparator	**Relative**
Sc-46	8.62	7.24
Ta-182	0.53	0.44
Fe-59	3782.71	5012.82
Mn -54	0.06	0.08
Co-60	0.03	0.02
Cs - 134	0.01	0.03
Cr-51	0.48	0.51
Zr-95	5524.53	6405.47
Zn-65	21328.59	20483.76
Na- 24	5.21	5.47
La-140	1.82	1.39
As-76	64.43	51.75
Al-28	265372.42	283183.81
Ti-208	2825.78	3223.98
Cs-138	48.53	52.43
Mg-27	53.91	50.5
Mn-56	152.62	171.9978
Cl-38	1.35	1.22

There are two main methods used to calculate isotopes concentration by using Neutron Activation Analysis (NAA) technique namely, relative and non-

relative method. Non-relative methods include absolute, comparator and K_0. At this study comparator and relative methods were used. In the NAA concentrations calculations of Comparator and Relative methods are close as shown in the table (4-15).

Table (4.16); Concentrations calculations of topaz as irradiated by (n+γ)

Isotope	A_e	m_e	A_s	m_s	Specific activity (A_e)	Specific activity (A_s)	C_s (ppm)	C_e (ppm)
Sc-46	3338.58	0.55	64.84	0.14	6094.75	4.48E+02	7.24E+00	9.86E+01
Co-60	4.98	0.55	13.78	0.14	9.09	9.51E+01	2.00E-02	1.91E-03
Ta-182	39.78	0.55	31.09	0.14	72.62	2.15E+02	4.40E-01	1.49E-01
Fe-59	21.8	0.55	1.85	0.14	39.79	1.28E+01	5.01E+03	1.56E+04
Mn-54	4.03	0.55	2.05	0.14	7.36	1.42E+01	8.00E-02	4.16E-02

Table (4.17); Comparison between concentrations calculations of topaz as irradiated by neutrons and irradiated by neutrons after γ irradiation

Isotope	Topaz irradiated by neutrons	Topaz irradiated by neutrons after γ irradiation
Sc-46	7.24	98.6
Co-60	0.02	00.0191
Ta-182	0.44	0.149
Fe-59	5012.82	15600
Mn-54	0.08	0.0416

The Concentration of (Sc-46) at topaz as irradiated by neutrons after γ irradiation is equal to that at Topaz irradiated by neutrons about "13" times while it is equal to (Co-60). (Ta-182) at the first sample is "3" times the second sample while the opposite for (Fe-59) and finally (Mn-54) at the first sample is twice the second sample.

Table (4.18); Concentration of elements (ppm)

Isotope	Isotopes concentration (ppm)		
	Reference material	**Topaz irradiated by neutrons**	**Topaz irradiated by neutrons after γ irradiation**
Sc-46	7.24	22.69436	98.57566
Co-60		0.008525250	0.001911272
Ta-182	0.44	0.4647348	0.1488706
Fe-59	5012.82	20838.70	15619.93
Mn-54	0.08	0.1426426	0.04158661

4.11.4. Transportation

4.11.4.1. Condition of transport of previously irradiated stones abroad [88]

The transport of radioactive material must be subject to a radiation protection program, which must consist of systematic arrangements aimed at providing adequate consideration of radiation protection measures. The nature and extent of the measures to be employed in the program must be related to the magnitude and likelihood of radiation exposure**.**

The following activities are measured one year after long irradiation in the ETRR-2 for 8 hours and became ready for transportation, where IATA [84] does not permit transportation for gemstones with activity more than 74(Bq/g).

Table (4.19); Activity of Topaz Sample before Shipment

Isotopes	T½ (day)	Bq/g
Ta-182	114.4	40.1
Mn-54	312.1	8.7
Sc-46	83.8	4.24
Zn-65	243.0	1.89
Cs-134	754.0	0.39
Sb-124	60.0	0.09
Co-60	1924.0	0.07
Ag-110m	249.0	0.03
Co-58	70.9	0.01
Total Bq/g	**55.52**	

The main residual radioactivity is due to Sc-46, Ta-182, Mn-54, Cs-134 and other isotopes which are equal to about 56 (Bq/g) which represents 75.67% of the IATA permissible level. Thus it could be transport without any problems [28].

Table (4.20); Some stones activities for one Kg in (Bq/g)

Bag	(Bq/g)
Sc-46	13.5
Mn-54	3.6
Fe-59	0
Cs-134	1.5
Ta-182	26.9
Total Bq/g	45.6
Total Bq	0.38 E6

For bulk measurements, one Kg of irradiated topaz stones of a very high dose rate and by selection and removing some stones of higher than the normal level for the reset stones, the dose rate drops gradually by removing stone by stone until goes to the normal level in the range of (4 μSv/hr).

Table (4.21); Some stones activities for another Kg in (Bq/g)

isotope	Bq/g
Sc-46	71.24
Mn-54	3.51
Fe-59	0.39
Cs-134	3.64
Ta-182	231

Table (4.18) and (4.19) are for the bulk. They clear that the isotopes activity ratios are differ from one Kg to the other.

4.11.4.2 Transport Index (Ti)

Transport index for a package means: the number expressing the maximum radioactive dose rate at distance of 1 meter from the external surface of the package.

Or;

The maximum radiation level in microsieverts per hour (μSv/h) at 1 m from the external surface of the package, divided by 10.

Example: 1μSv/h = 0. 1 mrem/h or Ti = 0. 1

Table (4.22); Transport Index (Ti) for bulk

No.	Bag	Maximum radiation level at any point on external surface	Transport Index (TI)	Category
1	Bag - 1	4.5 µSv/hr= 0.45 mrem/h	Ti = 0. 45	I-White
2	Bag – 2	3.0 µSv/hr= 0.3 mrem/h	Ti = 0. 3	I-White
3	Bag – 3	2.0 µSv/hr= 0.2 mrem/h	Ti = 0. 2	I-White
4	Bag – 4	3.3µSv/hr= 0.33 mrem/h	Ti = 0. 33	I-White
5	Bag – 5	4.7 µSv/hr= 0.47 mrem/h	Ti = 0. 47	I-White
6	Bag – 6	4.1 µSv/hr= 0.41 mrem/h	Ti = 0. 41	I-White
7	Bag – 7	2.8 µSv/hr= 0.28 mrem/h	Ti = 0. 28	I-White
8	Bag – 8	4.8 µSv/hr= 0.48 mrem/h	Ti = 0. 48	I-White
9	Bag – 9	4.9 µSv/hr= 0.49 mrem/h	Ti = 0. 49	I-White
10	Bag – 10	4.2 µSv/hr= 0.42 mrem/h	Ti = 0. 42	I-White
11	Bag - 11	4.4 µSv/hr= 0.44 mrem/h	Ti = 0. 44	I-White

** Measurements were done by G-M Probe FH-40.*

** Surfaces have no contamination.*

** Measurements were performed by Contamat - FHT 111 M.*

Figure (4.33); Topaz samples bags

The united Nations Number (UNN) or UN IDs are four-digit numbers that identify hazardous substances in the framework of international transport. Some hazardous substances have their own UN numbers while sometimes groups of chemicals or products with similar properties receive a common UN number. For the same material the number changes according to its state. Also it is differ dependent on levels of purity or concentration in solution. UN numbers range from UN0001 to about UN3500 and are assigned by the United Nations Committee of Experts on the Transport of Dangerous Goods. They are published as part of their Recommendations on the Transport of Dangerous Goods. These recommendations are adopted by the regulatory organization responsible for the different modes of transport. The (UNN) is displayed on the placards (lower half).

Table (4.23); List of some United Nation Numbers [85]

Number	Name and description
2910	Radioactive material, Excepted package.
2911	Instruments or Articles.
2909	Empty packages.
2912	Low specific Activity materials (LSA).
2913	Contaminated Object (SCO).
2977	Uranium Hexafluoride fissile (UF6)

The UN number for the bulk in this study is UN 2910 as illustrated at fig. (4-19)

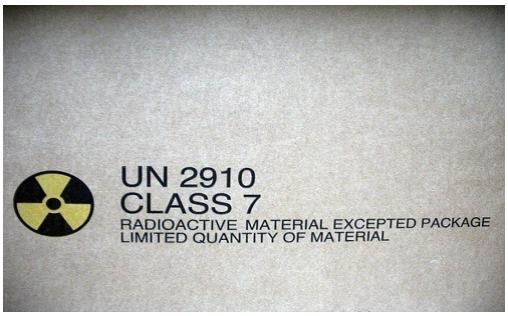

Figure (4.34); The UN number

4.11.5. Radiation doses

1. It is possible to calculate radiation dose from 500 kg of irradiated gemstones that contain exempt concentrations nuclides.
2. Using Micro Shield 5 (French code) and calculations using the appropriate gamma constants, we should see up to (5mR/hr=0.05mSv/hr) at a distance of 1 meter from a point source, soon after irradiation.
3. Actual measurements are much lower because of decay (<1mR/hr=0.01mSv/hr).

4.11.6. Storage of Radioactive Materials

Accidents can happen in all modes of transport, and it is likely that a significant number of accidents will involve packages of radioactive materials. Shielding packages are classified in three categories, I-White, II-Yellow and III-Yellow (table 2.6). Radioactive materials except those in category I- WHITE packages, shall be kept separated from living accommodations, from regularly occupied working spaces that may be continually occupied by passengers or the pubic.

By package is meant the packaging together with its radioactive contents as presented for transport. Packaging may, in particular, consist of one or more receptacle, absorbing materials, spacing structure, radiation shielding and devices for cooling, for absorbing mechanical shocks and for thermal insulation. There are six types of packaging. The one important at this study is the *Excepted Package* which is a packaging containing excepted radioactive material and designed to meet the general requirements for all packaging and packages [(116)].

Figure (4.35); Topaz bulk storage

4.11.7. Exclusion, Exemption and Clearance

Isotopes Half-lives in topaz by day are (83.81, 312.1, 44.51, 754.1 and 114.43) for (Sc-46, Mn-54, Fe-59, Cs-134 and Ta-182) respectively.

Table (4.24); Isotopes activity for bulk (with different weights)

Grams	Ta-182	Cs-134	Fe-59	Mn-54	Sc-46
988	86.36	0.35	0.15	1.92	32.75
1000	114.40	2.60	0.02	3.30	28.60
1015	85.43	0.57	0.09	4.64	42.57
1132	59.01	1.36	0.08	3.77	35
945	81.76	0.72	0.29	3.04	45.26
979	93.61	2.10	0.03	3.50	53.04
1010	92.56	1.34	0.10	3.08	52.81
1005	83.28	2.52	0.32	4.60	50.00
1091	51.79	2.51	0	1.91	40.02
1012	106.10	1.31	0.30	2.75	42.15
1015	122.23	0.90	0.33	4.10	43.52
1132	71	1.14	0.33	3.6	37.05
1029	44.17	11.75	0.17	1.9	45.88
920	100.00	1.00	0.18	5.34	48.20
1091	41	5.4	0.07	4.1	57.31

Treated gemstones should be deal as Building materials which exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at the most. This is the excess gamma dose to that received outdoors [85].

4.11.7.1. Determining exemption concentrations

1. The licensee will characterize every gemstone with respect to radionuclide ID and concentrations.
2. This would involve performing gamma spectroscopy on every gemstone.
3. In reality, this is nearly impossible because of the very large number of gems, the cost of gamma spec equipment, and lack of HP training.
4. Luckily, the NRC does not seem insistent on this point, and seems willing to accept a sampling plan.

Small gemstones, whether mounted or not, are not exempt from any disclosure requirements [12]. A licensee is not required to label ccontainers when they are in transport and packaged and labeled in accordance with the regulations of the department of transportation [117].

Counting equipment must be carefully calibrated if minimum detectable levels in topaz are to be determined. Minimum detectable levels are important if exempt concentrations, as listed in 10 CFR 30.70, are to be determined in irradiated topaz. In this study, irradiated gems were counted singly on all radiation equipment. Sampling large batches of irradiated topaz to determine average concentration levels (nCi/g=37 Bq/g of gem) does not ensure that individual gems will not occasionally exceed an exempt concentration level because the majority of activity, for any particular nuclide, could be concentrated in one or a few stones [9].

On june25, 1986, the (NRC) issued a letter to all non-power reactors (NRC, 1986) [118] that stated in part that "the distribution of irradiated materials, even with low levels of induced radioactivity, to unlicensed person is prohibited

unless the distribuer of such materials has a specific license, which permits such distribution. The staff considers gems to be adornments, and has not granted licenses for the distribution of irradiated gems. If the distribution of irradiated products to unlicensed persons, it must to obtain a license to reflect this activity. "This regulation or law has been in effect for many years but was ignored because after decay, neutron irradiated gemstones reached background levels [(2)]. the inequity in the irradiated topaz trade for gem dealers in the (USA) is that the exempt concentrations accepted by the (NRC) for the various radionuclides are those listed in 10 CFR 30.70, which are based on ingestion or inhalation and have not been a adjusted for solid, nonsoluble and noneatable (or digestible) materials such as gemstones that remain outside the human body. The (NRC) even requires that license applicants ask for an exemption to 10 CRF 32, 11 (C) since they "may consider gems to be products intended for application to human beings (NRC, 1988) [(119)]. The DOT Regulation 49 CFR 173,403 (X) states: "Radioactive material means any material having a specific activity greater than 0.002 microcuries per gram (2 nCi /g)." It concern to exempt concentrations for the release of irradiated gemstones [(120)]. the applications for exemption of very low level radioactive material will be judged by the whole body dose equivalent numerical criteria of 10 mR/person/year for the most exposed individual, or 1 mR(1 mR=0.01 mSv)/person/year for a large group and 1000 man Rem/year collective dose to the entire population [(121)]. For treated gemstones by reactor neutrons the level did not exceed 2 nCi/g. the total average whole body dose equivalent is about (3.6 mSv) per year or 1 mrem per day. (2 nCi /g) is no thing comparable to occupational dose rate or even to natural radioactive from natural gemstones which is higher than the treated gemstones by almost 1700 times, but natural gemstones are not subject to any regulations because it occurs naturally. Moreover the human body contains about 200 nCi (7400 Bq) of ^{40}K and nearly 100 nCi of ^{14}C.

4.12. Final Conclusion

1. Topaz (and some other gems) may be irradiated to cause it to change color.

2. Irradiation can induce radioactivity, albeit in exempt concentrations.

3. Irradiated gems exhibit only low levels of radiation and do not pose a health risk.

4. The color of topaz natural or irradiated by γ-rays easy to be removed or deformed if the stones suffered from temperature higher than 450 C^{o} while the stones irradiated by neutrons still stable without any deformation under highest temperatures treated.

5. The topaz used at this study is mainly used as gemstones, for scientific and technological applications.

6. To be irradiation for the treated gemstones production in the reactor with acceptance feasibility it must be irradiate among other activities and does not run the reactor especially for it; to save fuel, money and time.

7. Dependent on experimental observations the optimum way to obtain best irradiated topaz by the lowest cost it is recommended to irradiate it first by γ then by neutrons at the reactor , will needs only short irradiation time at this case, and finally irradiate for very short time in accelerator to give it its attractive shape.

8. Several elements were determined from these samples with different concentrations and compared with other works.

9. Trace element levels in gemstones affect their color, beauty and thus their market value.

10. In particular elements such as Sc, Ta, Mn and Fe are especially significant to the activities at these gemstones irradiated by neutrons.

11. Irradiation and ion implantation improve characteristic of minerals used at the study.

12. It was interesting to observe the analogy between topaz and quartz. Some defects could be similar in both materials.

13. Irradiation is used on gemstones to affect a variety of color changes; these colors are stable to light and heat in the neutron irradiation case, and the same behaviour for gamma irradiation but heat to finite range.

14. Blue topaz is still the most common gemstone being irradiated at the market.

15. AD in all, topaz is one of our least expensive precious gems that are still in high demand because of its intrinsic and enhanced beauty.

16. The treated gemstones cannot be distinguished from its natural by gemmological testing at this time.

17. There is an apparent correlation between the irradiation time and the color of the stones; the longer the time the deepest a color.

18. The blue color at reactor topaz is not eliminated or reduced by annealing and remains stable to the maximum temperature used for this study.

19. By using inductively coupled plasma mass spectrometry with laser ablation, most elements in the periodic table may be analysed in gem stones samples at (ng / g) levels.

20. With (ICP-MS), gem stones samples can be rapidly and inexpensively screened for purity without any need to remove solid and liquid inclusions prior to analysis. Thus ICP-MS is a powerful technique for trace multi-element and isotopic analysis.

21. More gemstone could be produced from Egypt by irradiation technique and new kinds of gemstone irradiation may become to be technically or economically available, enlarging the gemstone irradiation service market.

22. The high efficiency of neutrons to induce defects in the lattice of treated gemstones was demonstrated by spectroscopic methods.

23. Annealing at high temperatures induces the loss of fluorine from the damage region. These changes reduce the optical absorption of the topaz in the range 2000-3000 nm.

24. Structural changes occurred after implantation and/or after thermal annealing leading to change on the characteristic of the treated gemstones.

25. Ion implantation of natural topaz produces an amorphous layer through the entire implanted region.

26. Ion implantation of natural beryl produces dark layer through the entire implanted region.

27. It could be suggest that for low concentrations of radionuclides with short half-lives which are induced in treated gemstones so, short storage of gems until the activity concentrations have decayed to inconsequential levels would eliminate any health risks associated with the manufacture, distribution, and use of such gems.

28. for radiological concerns:

a. No contamination is observed.
b. Radionuclide concentrations will likely be only a fraction of exempt concentrations.
c. Radiation dose and dose rates expected to be low, even for large quantities of gemstones**.**

29. for licensing

a. Need to be able to demonstrate that “hot "gems do not escape detection.

b. Need to be able to demonstrate that gems do not exceed exempt concentrations for any nuclides (the “sum of fractions" rule applies).

c. Need to demonstrate that the licensee can make these determinations to the satisfaction of regulators.

30. Prospective licensees must be able to convince the regulators that they are competent to determine that gems are exempt from regulation.

31. Raman spectroscopy is an effective tool in uncovering treated stones, which should assist in enforcing proper disclosure.

32. Use of advanced testing methods, the accurate identity of some treated gemstones can be difficult to establish. Advances in modern technology are

expected to produce new gem treatment methods. These in turn will require the ongoing development of new gem-testing methods and improved gem identification criteria.

33. Positron annihilation is a powerful tool for detecting defects in topaz samples as pure mineral and as a gem.

34. Commercial Factors

The supply of treated blue stones has remained strong since the early 1970s, when they were first identified in the marketplace, until the present. It might have been expected that the existence of the irradiation process would cause a significant increase in the value of colorless topaz but this has not happened, possibly for two reasons. First, the costs of cutting and irradiation are sufficiently greater than the cost of the colorless topaz itself, so that these steps appear to account for 90% or more of the entire cost of the blue topaz produced. Colorless topaz is suitable for treatment should any additional demand arise. Gemstone irradiation is carried out between such activities to reduce the cost.

4.13. The future work

1. Dealing the samples with heat treatment to very high temperatures (around thousand) for different times to study all effects.
2. Convert topaz from solid state to powder in grains of sizes from 0.75 to 0.15 mm.
3. Trying to use topaz as (TLD) at practical work; where it is very sensitive for radiation by study the TL emission of natural samples.
4. Scan all samples with full spectrum by "IR" at the range of (7500–400) cm^{-1} at different resolutions and show remarkable changes mainly in the regions more effective and illustrate the effect of thermal treatment at those regions.
5. Using K_0 – NAA analysis for all samples to calculate isotopes concentrations.
6. Use a collection of irradiated topaz by neutrons and gamma for different doses to study the defects carefully by positron annihilation technique.
7. Study the thermal stability of defects by (PAS) technique to clear if it is reduced or growth.
8. Investigate the response of treated samples and the thermal stability of the EPR lines with electron paramagnetic resonance (EPR) spectroscopy.
9. Irradiate large pieces of crystals with a high dose and measured by OA.
10. Study Changes of TL glow curves for treated gemstones after different types of irradiation.
11. Measuring the depth distribution of implanted ions in raw and irradiated samples for mineral used at the study, record it annealing before and with annealing. However, the depth distribution and simulate it by using code like SRIM.
12. Trying to use the SSEF Gem LIBS package for gem stones identification without any other tools.
13. Using SIMS to diagnose color changes in heat treated gems.
14. Measure the dielectric properties (dielectric constant, loss, conductivity).