Summary

Since the short-lived cyclotron produced radioisotopes are finding increasing applications in medicine, industry, research studies and other fields of life sciences, especially gallium-67, indium-111 and thallium-201, therefore the main objective of this work is to study the separation and purification of these cyclotron produced radioisotopes as well as recovery of the encountered highly expensive target materials.
This thesis consists of three basic chapters:

Chapter(1) Introduction:

This chapter includes the routine routes of radionuclide production by neutron and charged particles activation using both reactor and cyclotron facilities. Structure, cross section and nuclear data importance are also reported. General chemistry of the products radionuclides and their target materials are illustrated. It also implies a concise literature review including the nuclear medicine application, production, irradiation, cooling, radiochemical separation and quality assurance tests of the respective product radionuclides.

Chapter (2) Experimental:

This chapter includes the various chemicals, reagents, radioactive materials and instrumentation used in this study. The details of the experiments carried out in this thesis are also reported implying the method of preparation of the used inorganic ion exchanger resin; 12-molybdocerate (IV). Description of the distribution behaviour investigations using static and dynamic studies and the factors affecting them; including, nature and chemical composition of the exchange medium and the amount of the radiotracer are illustrated. The chromatographic column separation studies and the factors affecting them; including, types, concentration and flow rate of the eluent are described confidentially.

Chapter (3) Result and discussion

Includes the obtained results as well as their analysis and discussion. Detailed and systematic studies including the distribution behaviour; breakthrough characteristics and elution profiles for the target and the product radionuclide couples have been carried out. The distribution behaviour of the individual metal ions is determined as a measure of the corresponding batch distribution coefficient (K_d) values as a function of nature and composition of the equilibrating media (HCl and HNO_3) as well as the concentration of the radiotracer on the sorbent material dried at 50°C in shaker thermostat adjusted at 25±1°C using batch equilibrium method. The obtained distribution behaviour relationships (K_d values in ml/g vs. concentration of the equilibrating medium) are discussed on the base of the target and product chemical species which are expected to predominate in the equilibrating solution and their interactions with the molybdocerate(IV) matrix. The experimental conditions necessary for high target/product separation factors ($\alpha = K_{d\ (product)}/K_{d(target)}$) are elucidated for each couple. It shows that the distribution coefficient (K_d) values of different cations under investigation from nitric acid are higher than from hydrochloric acid solution. Also, the values are high at low acid concentration and decrease with the increase in acid concentration. As well, the distribution coefficient (K_d) values decrease with increasing concentration of the radiotracer. The corresponding uptake values for the individual metal ions are obtained by the column breakthrough studies. It shows that fast breakthrough of cations under investigations is observed the higher the concentration of the radiotracers. Also, fast breakthrough was attained with acid concentration increasing where metal ions in hydrochloric acid move along the column bed faster than in nitric acid solution. Breakthrough characteristics for each target and product radionuclide in different acid media have been included. The separation performance of each target/product couple has been investigated using small chromatographic columns of 1g 12-molybdocerate(IV) matrix by both elution and frontal separation methods. The elution performance, radioactivity concentration and elution yield of the product radionuclide from 12-molybdocerate(IV) column matrix are implemented by investigations under comparable conditions for nature, chemical composition and flow rate of the eluent effects. In elution chromatography, it is observed that the position of maximum elution peaks was displaced to higher eluate volumes with increasing the flow rate of the eluent and with nitric acid than that with hydrochloric acid solution. Quality control studies, including radiometric determinations and chemical analysis, proved that the obtained eluates are radionuclidic, radiochemical and chemically pure and satisfy the considered limits for use of such radionuclides in nuclear medicine. From the obtained results and its discussion we can indicate that the optimum operating conditions for separation of carrier-free gallium(III) from bulk amount of zinc(II) can be achieved by using small chromatographic columns (0.6 cm i.d x 3.5 cm) filled with 1g 12-molybdocerate(IV) gel and passing 5ml mixture solution consists of $5x10^{-4}$M zinc(II) and 10^{-5}M gallium(III) in 0.05M HNO_3. Elution and purification of the bed matrix from retained Zn(II) is carried out successively with passing 3ml 0.05M HNO_3 acid and 1ml 0.05M HCl acid solutions. Thereafter, gallium(III) is eluted and purified with 7ml 0.5M HCl acid solution at a flow rate of 1 ml/min. The elution yield of gallium(III) was found to be about 94.7% of the total radioactivity present on the column. Separation of carrier-free indium(III) from bulk amount of cadmium(II) can be achieved by loading 5ml mixture solution consists of $5x10^{-4}M$ Cd(II) and $10^{-5}M$ In(III) in 0.01M HNO_3 onto 1g 12-molybdocerate(IV). Elution and purification of the bed matrix from retained Cd(II) is carried out successively with passing 5ml 0.01M HNO_3 acid and 2ml 0.05M HCl acid solutions. Thereafter, In(III) is eluted and purified with 14ml 0.1M HCl acid solution at a flow rate of 1 ml/min. The elution yield of indium(III) was found to be about 97.3% of the total radioactivity present on the column. Separation of carrier-free indium(III) from bulk amount of silver(I) can be achieved by loading 5ml mixture solution consists of $5x10^{-4}M$ Ag(I) and $10^{-5}M$ In(III) in 0.01M HNO_3 onto 1g 12-molybdocerate(IV). Elution and purification of the bed matrix from retained In(III) is carried out successively with passing 10ml 0.1M HNO_3 acid and 4ml 0.2M HNO_3 acid solutions. Thereafter, Ag(I) is eluted and purified with 25ml 1M HNO_3 acid solution at a flow rate of 1 ml/min. The elution yield of indium(III) was found to be about 98.6% of the total radioactivity present on the column. Separation of carrier-free thallium(I) from its lead(II) can be achieved by loading 5ml mixture solution consists of $5x10^{-4}M$ Pb(II) and $10^{-5}M$ Tl(I) in 0.01M HNO_3 onto 1g 12-molybdocerate(IV). Elution and purification of the bed matrix from retained Pb(II) is carried out successively with passing 7ml 0.05M HNO_3 acid and 2ml 0.05M HCl acid solutions. Thereafter, Tl(I) is eluted with 18ml 1M HCl acid solution at a flow rate of 1 ml/min. The elution yield of thallium(I) was found to be about 92.4% of the total radioactivity present on the column.

Frontal separation method had been used for separation of pairs of target materials and produced radionuclides present in large solute volumes. So, the best condition for frontal separation of Ga(III) from its target Zn(II) is by passing 35ml mixture solution of $5x10^{-4}$M Zn(II) and 10^{-5}M Ga(III) in 0.05M HNO_3 through a chromatographic column packed with 1g 12-molybdocerate(IV) gel at a flow rate of 1ml/min. **Frontal separation** of In(III) from its target Cd(II) can be achieved by passing 60ml mixture solution of $5x10^{-4}$M Cd(II) and 10^{-5}M In(III) in 0.01M HNO_3 through a chromatographic column packed with 1g 12-molybdocerate(IV) gel at a flow rate of 1ml/min. Frontal separation of In(III) from its target Ag(I) can be achieved by passing 200ml mixture solution of 10^{-4}M Ag(I) and 10^{-5}M In(III) in 0.1M HNO_3 through a chromatographic column packed with 1g 12-molybdocerate(IV) gel at a flow rate of 1ml/min. Frontal separation of Tl(I) from its target Pb(II) can be achieved by passing 200ml mixture solution of $5x10^{-4}$M Pb(II) and 10^{-5}M Tl(I) in 0.05M HNO_3 through a chromatographic column packed with 1g 12-molybdocerate(IV) gel at a flow rate of 1ml/min.