

This can be explained on the basis of various factors, such as:

- (i) The type of fibers of the papers.
- (ii) The conditions of pulping and bleaching used during the manufacture of the papers.
- (iii) Type of sizing and loading materials.
- (iv) The nature of the chemical bonds between ink and paper.
- (v) Oxidative destruction and other transformation of ink dye.
- (vi) Resins and polymers in ballpoint inks.

It should be mentioned that the nature of the cellulosic fibers is a very important factor, which affects the stability of the aging inks on the papers. The fibers of yellow writing paper made from bagasse pulp and ruled writing paper made from straw soda pulp have accessible cellulosic fractions as resulted from higher percentage of pentosan and lower crystallinity which increases the water retention value (W.R.V) Table (1). These greater hydrophilic fractions of the fibers of yellow and ruled papers, as well as short time of sizability and lower thickness caused a higher penetration of the inks through the paper fibers and consequently rises stability of inks to aging process.

On contrast, the presence of rosin as sizing materials in the Japanese and Brazilian papers had the effect of filling the surface pores, leading to an increase in the time sizability (Table 9). In addition, the lower content of hydrophilic fractions of these types of papers decrease the penetration of the ink through the paper fibers and consequently lowers stability of inks.

However, the all types of papers used in this work contained fillers as CaCO_3 in case of Japanese, Brazilian and Ruled papers, and CaSO_4 in case of yellow papers, which increases the resistance of ink placed on the surface of paper to aging.

Figures (4-7) represents the aging curves of different types of inks on Japanese, Brazilian, Ruled and yellow papers respectively. It's clear from figure (4) that the blue ballpoint ink model Bic and iron blue liquid ink on Japanese paper have nearly the same stability to aging, but both types are higher than the stability of blue ballpoint ink model Reynolds. This can be reasonably understood by assuming that the aging process of the inks on the paper surface begins as soon as the ink is placed on the paper and the volatile components evaporate, leaving the dyes and resins adhering to the writing surface or penetrate through the paper fibers depending upon the type of the fibers, sizing material and filler. By prolongate the time of aging, oxidation of iron compound and dye takes place in case of iron blue liquid ink while oxidative destruction and other transformations of ink dyes, resins and polymer in blue ballpoint ink occurred⁽²³⁾, either on the surface of the paper in case of well sized paper (Japanese paper) or through the intermolecular spaces of the paper fibers in case of other less sized papers. Although the iron blue liquid ink had ability to penetrate slightly, the well sized Japanese paper due to its very lower viscosity than ballpoint inks leading to lower extraction with solvent (more stability).

On the other hand, the greater oxidation and cross-linking of ingredients of ballpoint ink model Bic with aging time, and the presence of chelated metalized copper phthalocyanine dyes lead to nearly the same stability to aging on this type of paper. However it is clear from the same figure that the blue ballpoint ink model Bic is more stable than the blue ballpoint ink model Reynolds towards the environmental conditions and chemical erasures used in this work. The presence of chelated metalized copper phthalocyanine dyes in ballpoint inks assisted stability of the ink more than the iron blue liquid ink.

Figure (5) shows that the blue ballpoint ink model Bic was more stable on Brazilian paper than the iron blue liquid ink up to 200 minutes aging time. By prolongation of the aging time to 400 min., it was found that no significant change with aging time takes place and the curves became level off. This is probably due to that fact that the prolongation of aging time will lead to an increase in hydrogen bonding and hence formation of contracted chains of the paper. The relative stability of all types of inks to aging increase or decrease due to oxidation of the ingredients of the inks inside the paper. At aging time 300 and 400 minutes the solvent extracted ink components, only from the outside layers with equal amount and cannot be extracted from the inner layer of the paper, specially iron blue liquid ink. Figure (5) showed also that the stability of the previous inks at all time of aging was higher than the stability of blue ballpoint ink model Reynolds.

Inspection of Figures (6) and (7) show that the stability of different inks on ruled and yellow papers obeyed the following order: Iron blue liquid ink > blue ballpoint ink (Bic) > blue ballpoint ink (Reynolds).

Thus, the liquid ink had the higher stability on ruled and yellow papers than blue ballpoint ink. This can be explained on the basis of the following reasons:

- (i) The fibers of these types of papers are composed of higher content of short-chain hemicellulose fractions which are readily wetted by liquid ink with lower viscosity. A result of surface wetting and capillary forces, the liquid ink had a higher ability to penetrate the less sized and thickness papers and become more stable to aging process.

- (ii) The presence of iron element in blue pigment of liquid ink resulting in a more complex formation (iron gallotannate) which associates to some extent inside the less sized papers and oxidized during the aging leading to more stability to aging.

It was shown from figures (4 –7) that different papers caused different shaped ink aging curves, which means that only different types of inks on the same papers can be compared for age, and the question then is to determine if the two curves can be related so that one curve can predict the other.

In forensic examination of handwritten documents, it is very important to ascertain whether a sentence or word of the text was written before or after the signature on a receipt or document if written with the same ink, and also to compare between suspected and legal words of the text.

Thus by comparing the rate and extent of extraction of absorption curves in the visible region, one can know which ink is oldest than the other on the same type of paper. Inks on the paper samples must have been stored under the same conditions (heat, light, humidity, and so forth).