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A Study of the Leachability and Solubility of some Radionuclides in the Waste Repository

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The safety of radioactive wastes in the repository depends on its capacity to confine radioactivity from being released to the biosphere. The process that most likely process can lead to the release of radionuclides from a repository is the transport by groundwater. The release rate of radionuclides from the solidified blocks is controlled by the solubility limits and leaching rate of radionuclides. The present work aims at investigating the solubility and leaching processes and their effect on the safety assessment of a radioactive waste in the repository. This work studies both processes with different concentrations of elements. The experimental study focused on the elements; strontium (Sr), cobalt (Co) and iron (Fe). To simulate the effect of radiation emitted from the radionuclides of these elements, the cement blocks that confine the elements are irradiated by Gamma (ɣ) Rays of doses; 40, 120, and 240 kGy. The results of non-irradiated and irradiated blocks are compared. The results indicate that ɣ radiation has a significant effect on the pH of the leachate. The relation between leaching rate and solubility is illustrated.

Keywords: The safety of repository, Chemical reaction, pH, Radiolysis, Leaching, Solubility

Introduction

The safety barriers in the repository consist of the waste immobilization media, concrete packages, the vault structure, the backfill material and geological formation (Nirex, 1993d). These barriers intend to provide a level of containment for a period of time to allow the radionuclides to decay to negligible levels of activity (Atkinson et al., 1993) [1].

After some time, in some cases, the containers in the vaults can have partially degraded/corroded areas to the point that water flow through the containers will be possible [2]. The chemical processes are the main factor in the degradation of waste and engineering materials. Therefore, neglecting chemical reactions in the safety assessment calculations of a disposal site is not safe in all cases. The chemical processes that are significant to the release and transport of contaminants in the near filed have been identified through a previous research and assessments [3]. It has been considered that the important processes concerning the chemicals that act as an indicator of chemical reaction of the near filed are: pH, leachability, sorption, and solubility. Sorption of elements, however, has been studied in a previous study [4].

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Leaching is the washing process of soluble salts and removal of elements through a porous material by the percolation of water [5-7]. The process of leaching depends on several chemical and physical factors and occurs when a liquid phase percolates or is in contact with a solid material [8,9].

The solubility of a material, on the other hand, depends on the physical and chemical properties of the solute and solvent as well as on temperature, pressure and presence of other chemicals (including changes in the pH) of the solution [10-18].

On the other hand, the solubility of a solute is the maximum amount of solute that can be dissolved in a certain amount of solvent or solution at a certain temperature [19].

The solubility and leaching processes of radionuclides affect directly the source term analysis of radionuclide transport that leads to risks to the human environment. To license a repository, it is essential to have confidence in many aspects of license conditions and the associate safety assessment [20].

The present work aims at investigating the effect of the leaching and solubility processes on the calculation of the source term of different elements' concentrations. Sr, Co and Fe are selected to be the elements investigated in this present study. Moreover, the cement blocks that confine the elements are irradiated by gamma rays to simulate the effect of radiation on the leaching and solubility processes.

II-Materials and Methods

Preparations of cement blocks

The cement material is mixed with different elements to form blocks. All experiments used cement blocks with water to cement ratio 0.6, the mixture (cement and solution (water+ salts)) is stirred for 1 minute and casted in cubic moulds of dimensions 0.8 cm× 0.8 cm× 0.8 cm. The blocks are removed after one day and left for a curing time of 28 days.

Experiment:

a- Leaching tests:

The tests were carried out on cement blocks containing Sr, Fe, Co separately and a a mixture form. The used concentrations are 0.01, 0.02, 0.1, and 0.15 M/L. The tests were carried out as follows:

1. The cement blocks were immersed in 300 ml of distilled water.
2. The pH values are measured before and after immersion of cement block in distilled water.
3. The leachate is collected and the key elements are determined after 90 days,
4. Calculate the leaching rate for the radionuclides after 90 days using the equation:

$$\text{Leaching Rate} = \frac{\text{Volume of Leachate} \times C_i}{T \times \text{Surface area}}\ g/m^2.hr$$ [21]

Where:

C_i: is the concentration of radionuclide in water, T: is time of experiment.

5. The other cement blocks are placed in 300 ml for 14 days,
6. The cement blocks were exposed to gamma rays at different doses as (40, 120, and 240 KGy),
7. All the steps were repeated with the irradiated cement blocks.
8. The solubility values were calculated for the radionuclides after 90 days for non irradiated and irradiated cement blocks.

III- Results and Discussions

A- *Chemical analysis*

Figure (1) shows the leaching results of Ca ions in cases of cold and irradiated cement blocks in the pure state and with additives (Sr, Fe, Co, and mixture salts).

The results indicate that calcium release increases when the cement blocks are irradiated. This may be due to the carbonation in the cement blocks. Bouniol & Bjergbakke show that the calcium hydroxide equilibrium can be affected by the radiolysis products releasing calcium ions to react. In the presence of hydrogen peroxide, calcium peroxide octahydrate is formed [22]. However, the released calcium ions can react with a wide range of chemical elements which can be leached from the cement matrix by the groundwater.

In irradiated cement blocks including Sr, Co and mixture of salts, there is a possible depletion of Portlandite and an increase of Calcite quantity on the surface of the blocks. This process, called carbonation, can be explained by the reaction of

Portlandite with the carbon dioxide present in the atmosphere, as shown in the equation below [22]:

$$Ca(OH)_2 + CO_2 \longleftrightarrow CaCO_3 + H_2O \quad \text{(Eq. 01)}$$

Figure (2) shows the leaching results of Na ions in all cases of cold and radiation experiments with the different cement blocks (pure cement, cement with Sr, cement with Fe, cement with Co, and cement with a mixture of salts).

In all cases of cement with the different additives (pure cement, Sr, Fe, Co, and Mixture salts) the results show that in case of cold experiment, there is a high release of Na ions, but in case of radiation, Na exhibits a low release as appeared in the results of the chemical analysis. It may be due to the formation of calcite which can block the cement pores and prevent the carbon dioxide penetration to the inner part of the cement blocks.

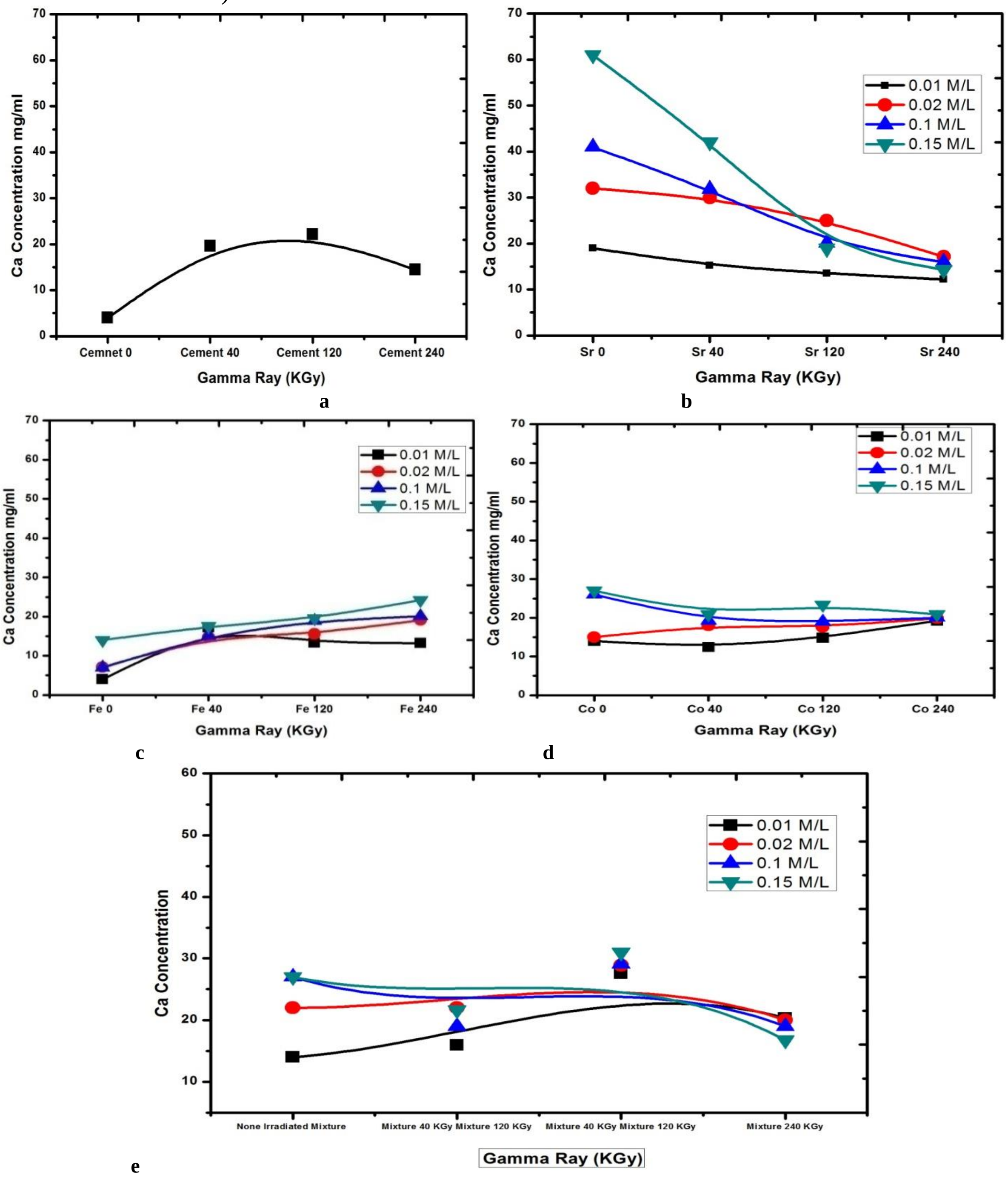


Fig. (1) Chemical analysis of Ca leachate after 90 days for cold and irradiated cement blocks at different doses

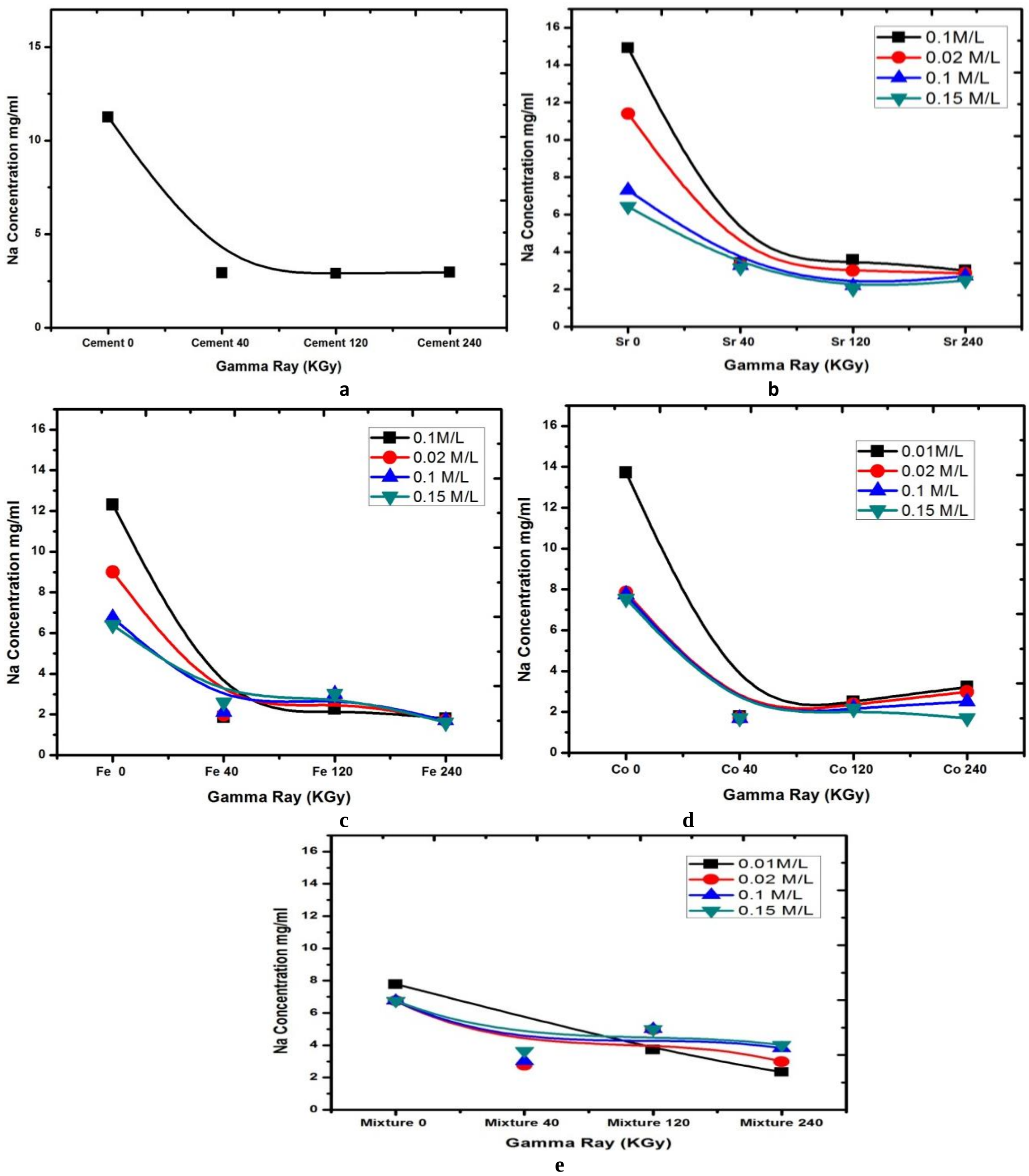


Fig. (2): Chemical analysis of Na leachate after 90 days for cold and irradiated cement blocks at different doses

Figure (3) shows the leaching results of Al ions in all cases; cold and radiation experiments with the different cement blocks (pure cement, cement with Sr, cement with Fe, cement with Co, and cement with mixture of salts).

In all cases of cement with the different additives, the results show that in case of cold experiments, Al has a high release. In cases of radiation, Al has a low release as appeared in the chemical analysis. It may be attributed to the reaction of Al with alkaline metals in the cement at a low pH value. This reaction takes place at the pH values in the range of 4.5-8.5 resulting in formation of a protective, highly adherent oxide film on the surface [23].

$$2Al + 6\ MOH \text{---} M\ [\ Aluminates] + 3H_2 \quad \text{(Eq. 02)}$$

Where M is Na, K, or Ca

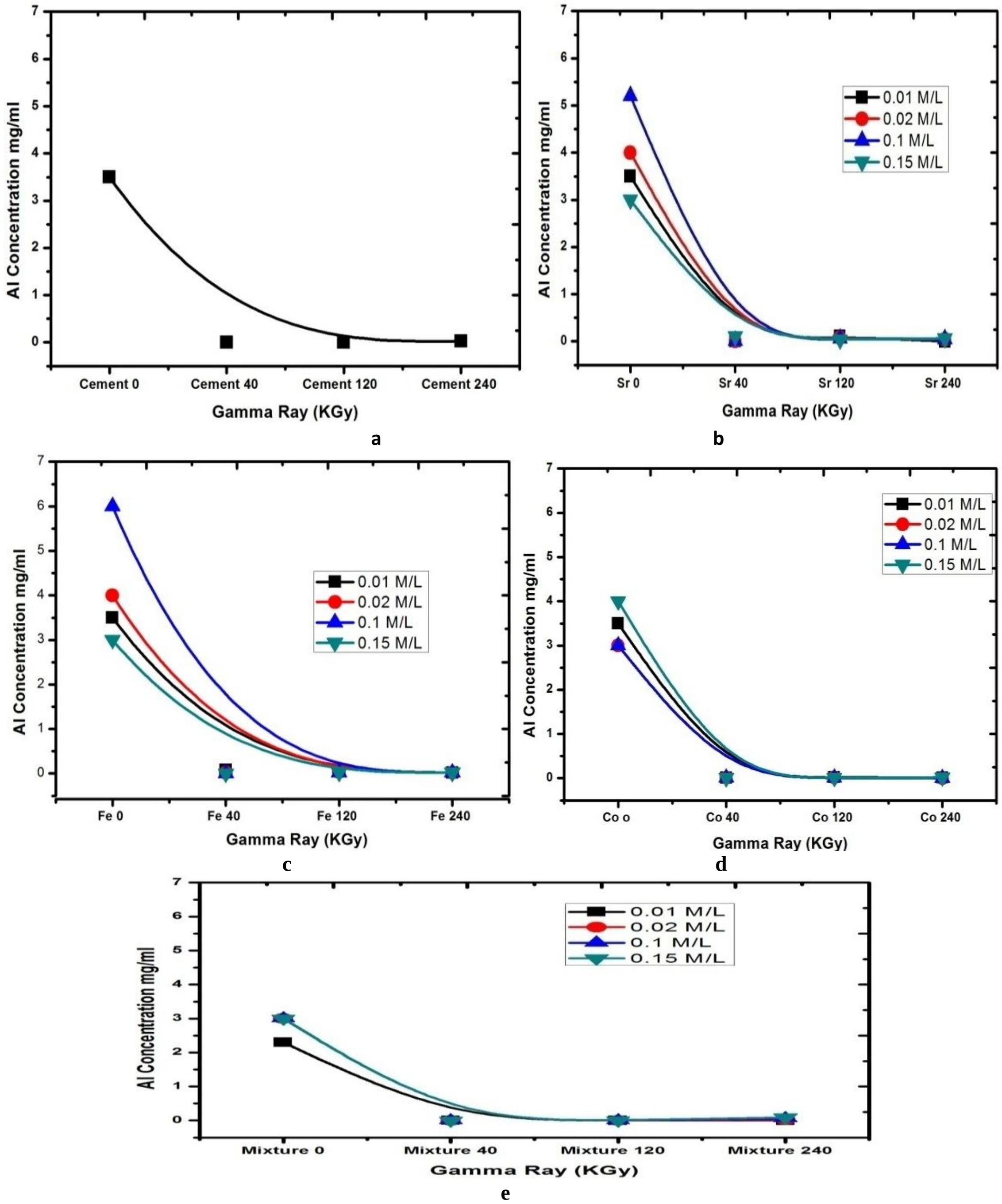


Fig. (3): Chemical analysis of Al leachate after 90 days for cold and irradiated cement blocks at different doses

B*- Leaching tests*

Figure (4) illustrates the change in pH values of non-irradiated cement and irradiated cement; first, pure cement blocks, second cement blocks and cement of the different concentrations of the elements are used, third cement blocks containing mixture salts.

Figure (4 a), shows that the change in the pH values of non-irradiated cement (pure cement and cement containing different salts) at different concentration of elements. Zero is the pure cement without additives, in the pure cement, the pH values reach to 12, it is fact about the cement when, it contacts with water the Ca $(OH)_2$ is released to the leachate, and some cations. It is may be the reason for the increase in the pH value. Strontium results in the highest pH value 14.0 and Cobalt gives the lowest pH values 12.5. The increase in the pH values for Sr^{+2} can be explained by the release of Sr^{+2}, Ca^{+2}, Na^{+1}, Si^{+4}, Al^{+3}, Cl^{-1}, and Fe^{+2} ions into the solution. The high concentration of Ca $(OH)_2$ in the solutions may be due to the replacement of Ca^{+2} by Sr^{+2}, which offers more pores filled by water. The presence of these ions Sr^{+2}, Ca^{+2}, Na^{+1}, Si^{+4}, Al^{+3}, Cl^{-1}, and Fe^{+2} in the solution increases its alkalinity [24-27]

$$\text{Ca-C-S-H} + Sr^{+2}_{(aq)} \leftrightarrow \text{Sr-C-S-H}_{(s)} + Ca^{+2}_{(aq)} \quad \text{(Eq. 03)}$$

Figures (4 b-d), illustrate the effect of gamma radiation on the leachability of cement blocks, either pure cement or cement containing chlorides of elements separately or a mixture. It is clear that the leachate pH values are generally lower than these for the non-irradiated blocks. The results also indicate the studied values of different doses (40, 120, and 240KGy). The differences between the pH values are very small. It can be seen that the effect of the concentration on the pH values for irradiated cement is negligible. However, for the dose 120 KGy, an increase in the pH values occurs for Sr at a concentration of 0.01 M/L. This can be attributed to the appearance of Sr due to radiolysis. The decrease of the pH values may be due to the radiolysis of water present in cement paste when interacts with gamma rays. The radiolysis leads to some products which can be highly reactive such as electrons, hydroxyl radicals and hydrogen peroxide. These radiolysis products will interact with cement paste components and its hydration products forming a wide range of compounds [16-18]. Bouniolet.al. stated that when the alkaline medium in concrete pores undergoes radiolysis, H_2 gas is produced as a primary product [22].

Interaction of gamma radiation with water results in radical ions and free sub-excitation electron as expressed by the equation [28].

$$H_2O \xrightarrow{rad} H_2O^{.+} + e^- \quad \text{(Eq. 04)}$$

C- Calculation of leaching rate

Table (1) lists the calculated leaching rates for non-irradiated and irradiated cement for the different cement blocks (cement with Sr, cement with Co, and cement with a mixture of salts).

The results indicate that, in case of Sr and Co, there is an increase in the values of the leaching rate for non-irradiated Sr cement blocks at the single state then the mixture of salts. It can also be seen that the leaching rate decreases when the cement blocks are irradiated. However, increasing the dose did not result in more effects. The decrease in the leaching rate by radiation may be attributed to the blockage of the pores of the cement leading to a decrease in the release of radionuclides.

Figure (5) shows the leaching rate of Sr in the separate salts and mixture of salts for both cold and irradiated cement blocks at different doses and different concentrations.

The results show that there is a decrease in the leaching rate with the different doses for the separate salts and the mixture of salts, except for the single salt at concentration of 0.1M/L where the leaching rate increases with increasing the dose by an equal value. This can be explained by the radiolysis of the water in the pores of cement blocks. As a result, many reactions take place with the free radicals.

Figure (6) shows the leaching rate of Sr in the separate salts and mixture of salts for both cold and irradiated cement blocks at different doses and different concentrations.

The results show that in case of Co single salt there is an increase in the leaching rate when the radiation doses increase. However, in the mixture of salts, the behavior of Co is different; the leaching rate decreases at high radiation doses and increases for the doses of 40, and 120 KGy.

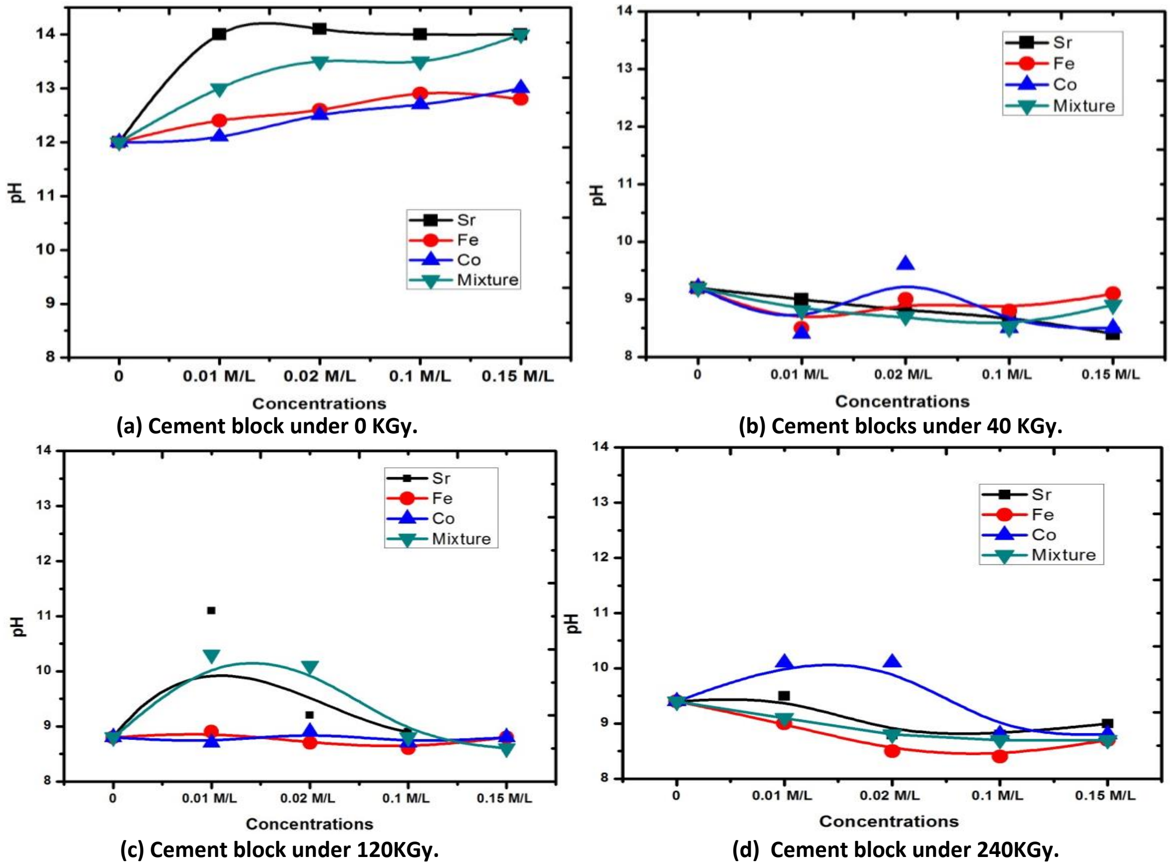
D- *Calculation of solubility values*

Table (2) lists the values of the Solubility values for cold and irradiated Cement blocks for different elements (Sr, Co, Fe, and mixture of salts).

The results show that there is a decrease in the values of solubility for Sr and Co as the effect of radiation in the single state and mixture of salts.

Figure (7) shows the calculated solubility values for both Sr single and mixture of salts in cold and irradiated cement blocks at different doses and different concentrations.

The results illustrate that the solubility decreases with the radiation for both single and mixture of salts, except for the concentrations 0.01, and at 0.02 M/L, it increases.



(a) Cement block under 0 KGy.

(b) Cement blocks under 40 KGy.

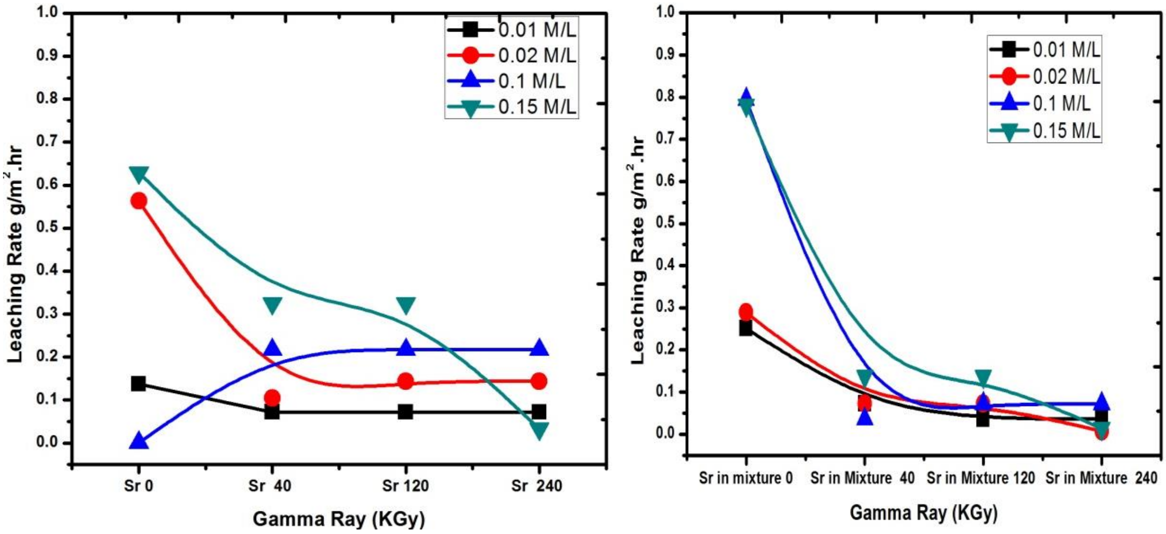
(c) Cement block under 120KGy.

(d) Cement block under 240KGy.

Fig. (4): The change in leachate pH of cold and irradiated cement blocks

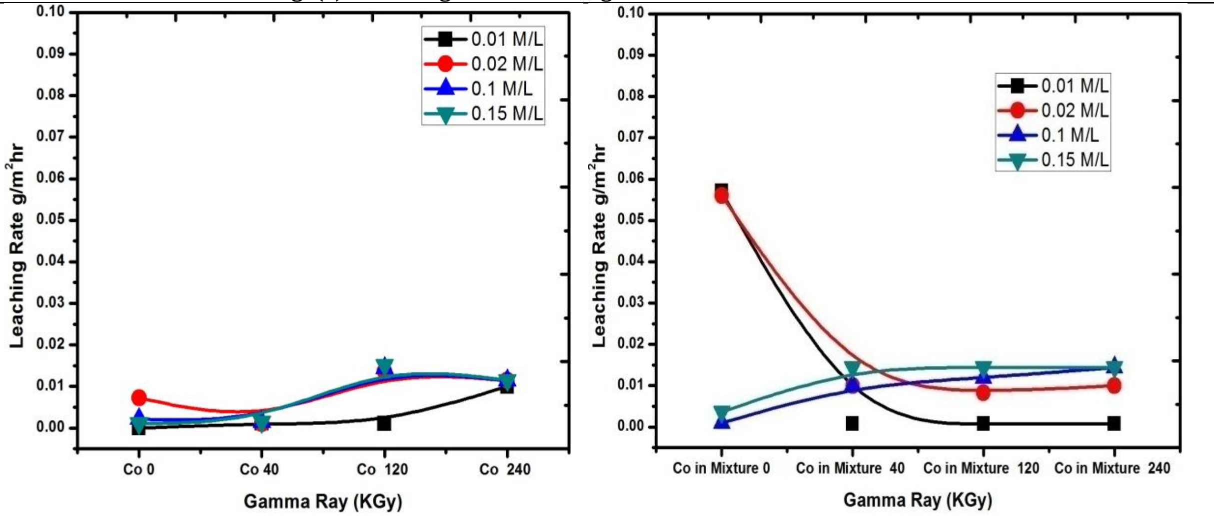
Table (1) The calculate of the leaching rate for cold and irradiated cement blocks

Radionuclides	0.01 M/L	0.02 M/L	0.1 M/L	0.15 M/L
Sr of None Irradiated	1.73 E-1	5.63 E-1	3.61E-3	6.29 E-1
Sr under 40 KGy	7.23 E-2	1.044E-1	2.17 E-1	3.25 E-1
Sr under 120 KGy	7.2 E-2	1.44 E-1	2.17 E-1	3.25 E-1
Sr under 240 KGy	7.2 E-2	1.44 E-1	2.17 E-1	3.25 E-1
Co of Non Irradiated	0000	7.23 E-3	2.17 E-3	1.085 E-3
Co under 40 KGy	1.085 E-3	1.085 E-3	1.44 E-3	1.44 E-3
Co under 120 KGy	1.058E-3	1.37 E-2	1.44 E-2	1.51 E-2
Co under 240 KGy	1.0 E-2	1.144 E-2	1.144 E-2	1.148 E-2
Sr in Mixture of Non Irradiated	2.51 E-1	2.89 E-1	7.95 E-1	7.81 E-1
Sr in Mixture under 40 KGy	7.23 E-2	7.23 E-2	3.6 E-2	1.37E-1
Sr in Mixture under 120 KGy	3.61 E-2	7.23 E-2	7.23 E-2	1.37E-1
Sr in Mixture under 240 KGy	3.61 E-2	7.23 E-2	7.23 E-2	1.37 E-2
Co in Mixture of Non Irradiated	5.7 E-2	5.6E-2	9.7 E-3	3.61 E-3
Co in Mixture under 40 KGy	7.23 E-4	1.0 E-2	1.0 E-2	1.44 E-2
Co in Mixtureunder120 KGy	7.23 E-4	8.31E-3	1.19 E-2	1.44 E-2
Co in Mixtureunder240 KGy	7.23E-4	1.0 E-2	1.44 E-2	1.44 E-2



a b

Fig. (5): Leaching rate for Sr single and mixture salts at different doses



a- b-

Fig. (6): Leaching rate for Co single and mixture salts at different doses

Table (2) The calculated solubility values for cold and irradiated cement blocks

Radionuclides	0.01 M/L	0.02 M/L	0.1 M/L	0.15 M/L
Sr None Irradiated	5.45E-6	1.13 E-5	1.77 E-5	1.98 E-5
Sr under 40 KGy	6.69 E-6	7.04 E-6	1.05 E-5	1.27 E-5
Sr under 120 KG	9.03 E-6	1.09 E-5	1.31 E-5	1.52 E-5
Sr under 240 KGy	8.35 E-6	9.57 E-6	9.78E-6	1.64E-5
Co None Irradiated	0000	3.39 E-7	1.018 E-7	5.09 E-8
Co under 40 KGy	5.09 E-8	5.09 E-8	6.78 E-8	6.78 E-8
Co under 120 KGy	5.09 E-8	6.44 E-7	6.78 E-7	7.12 E-7
Co under 240 KGy	5.09 E-8	5.59 E-7	6.95 E-7	5.09 E-8
Sr in Mixture None Irradiated	8.08 E-6	9.09 E-6	2.50 E-5	2.45 E-5
Sr in Mixture under 40 KGy	7.04 E-6	8.74 E-6	1.02 E-5	1.04 E-5
Sr in Mixtureunder 120 KGy	6.92 E-6	8.31 E-6	1.29 E-5	1.84 E-5
Sr in Mixture under 240 KGy	8.28 E-6	9.67 E-6	1.05 E-5	1.49 E-5
Co in Mixture None Irradiated	2.68 E-6	2.63 E-6	4.58 E-7	1.69 E-8
Co in Mixture under 40 KGy	3.39 E-8	4.92 E-7	5.09 E-7	6.78 E-7
Co in Mixture under 120 KGy	3.39 E-8	3.90 E-7	5.59 E-7	6.78 E-7
Co in Mixture under 240 KGy	3.39 E-8	5.09 E-8	6.78 E-8	6.78 E-8

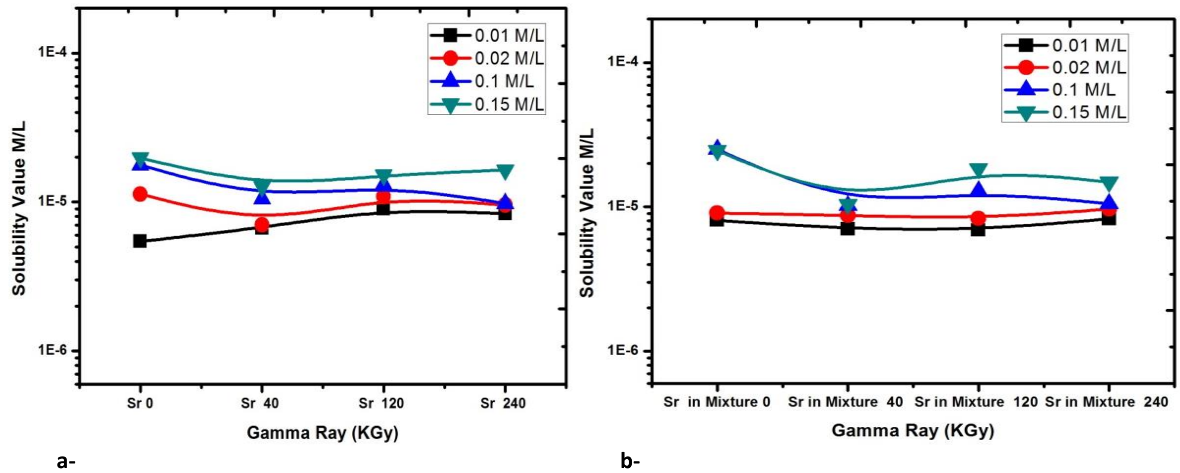


Fig. (7): Calculated solubility values for Sr single and mixture of salts with different doses

Figure (8) shows the calculated solubility values for both Co single and mixture of salts in cold and irradiated cement blocks with different doses at different concentrations.

Figure (8 a) shows that the solubility of Co at the single salt_increases with the radiation at the concentrations of 0.02, and 0.1 M/L. For the concentration of 0.01M/L, it undergoes no change, and decreases for the concentration of 0.15 M/L.

Figure (8 b) illustrates the case of Co in a mixture of salts. It shows that the solubility decreases with the increase in radiation doses for the concentrations 0.01, and 0.02 M/L. The behavior is different for the concentrations 0.1 and 0.15 where the solubility increases with the radiation doses 40, and 120 KGy then it decreases at the radiation dose of 240 KGy.

From the values of the leaching rate and the solubility values, a relation can be developed between the leaching rate and the solubility values by plotting the solubility as a leaching rate at the Y- axis and on the X- axis.

Figure (9) shows the relation between the leaching rate and solubility calculated values for Sr in the single salt at the different doses. From the figures, it can be seen that the relation is nearly linear for all doses.

Figure (10) shows the relation between the leaching rate and solubility calculated values for Sr in the mixture of salts with the different doses. The results of Sr in the mixture of salts also show that the relation between the leaching rate and solubility calculated values is linear.

Figure (11) shows the relation between the leaching rate and solubility calculated values for Co single salts at different doses. The results illustrate that the relation is linear for all doses and there is no effect of the radiation on the relation between the leaching rate and solubility calculated values.

Figure (12) shows the relation between the leaching rate and the solubility calculated values for Co in a mixture of salts.

In case of Co in a mixture of salts, the figures indicate that the relation between the leaching rate and solubility is linear also.

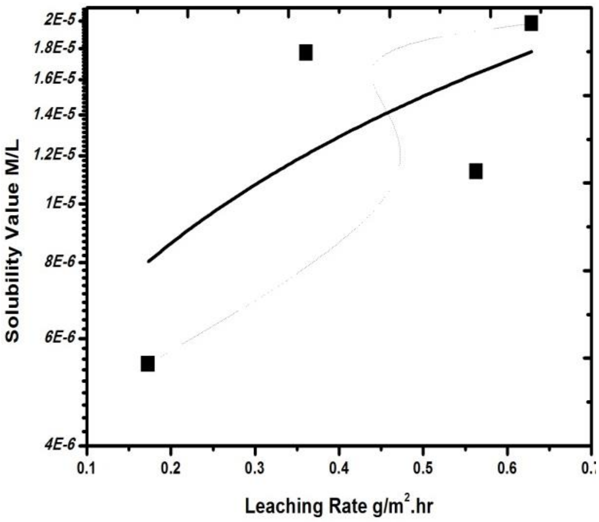
From these Figures, it can be seen that most of the relations between the solubility and leaching rate are linear. Thus, the linear equation could be used to find the relation between them from this equation:

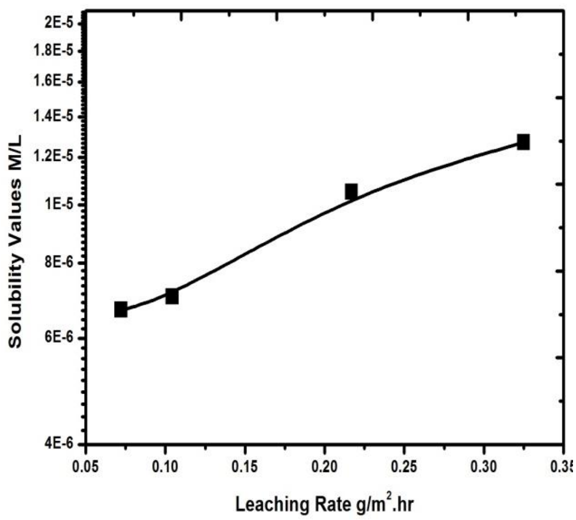
$$y = a + bx$$

Where,

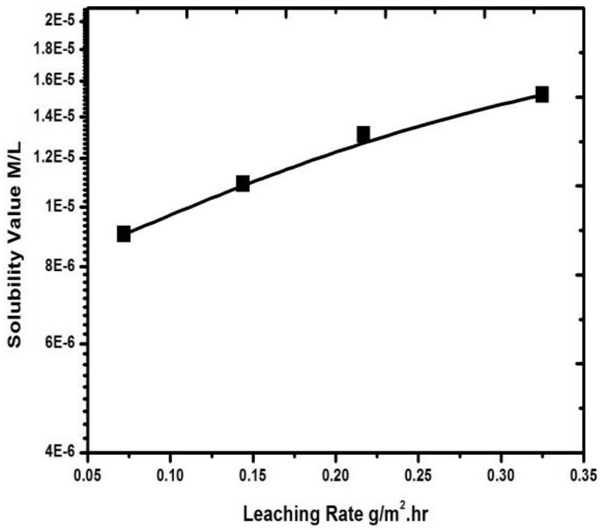
y: is the solubility, a: intercept, b: is the slope, x: is the leaching rate

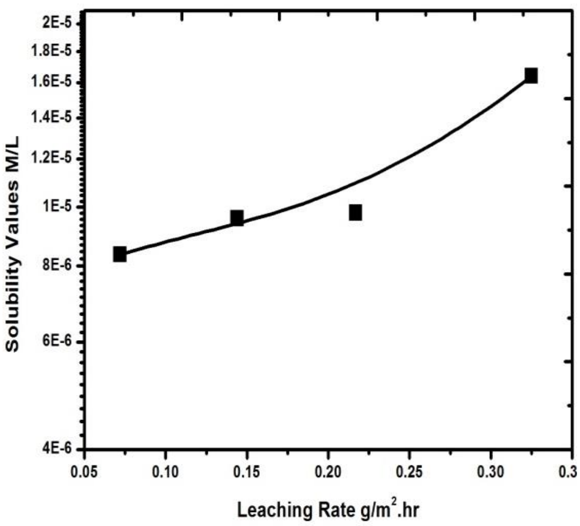
Table (3) lists the conditions and the percentages of solubility to the leaching rate. The results illustrate that the solubility of Sr and Co in the single and mixture of salts at both cases; the cold and irradiated cement blocks are nearly equal.





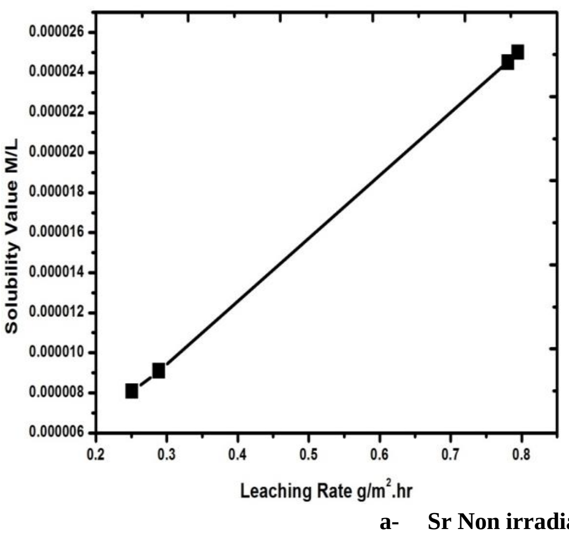
a- Sr Non Irradiated b- Sr under 40 KGy

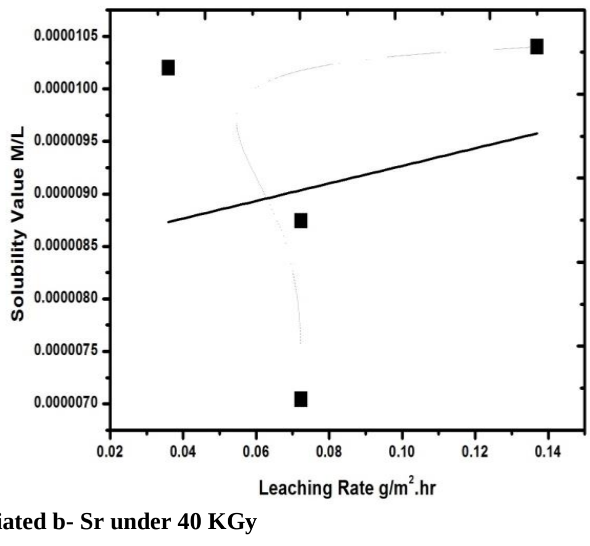




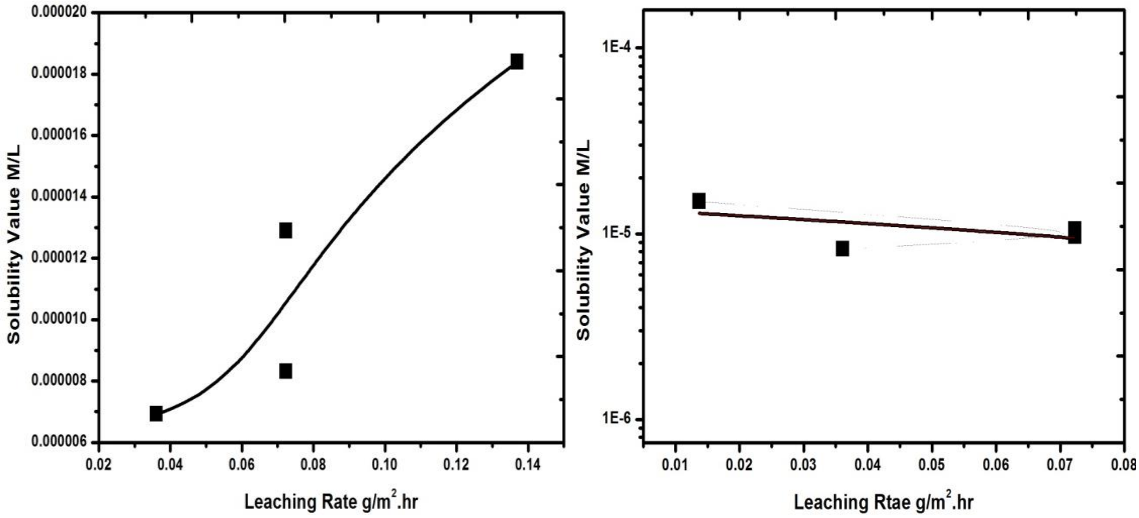
c- Sr under 120 KGy d- Sr under 240 KGy

Fig. (9): The relation between the leaching rate and solubility calculated values for Sr single salt at different doses



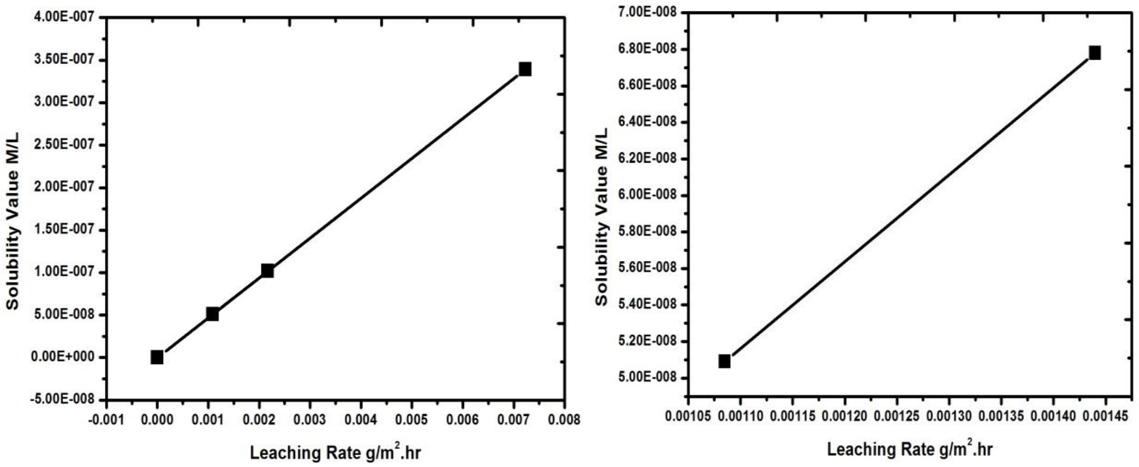


a- Sr Non irradiated b- Sr under 40 KGy

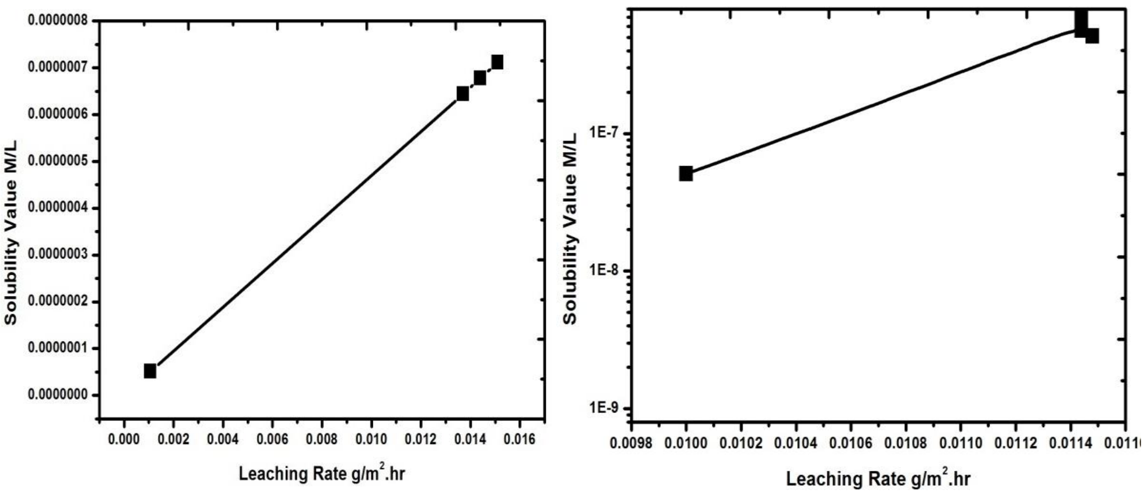


c- Sr under 120 KGy **d- Sr under 240 KGy**

Fig. (10) The relation between the leaching rate and solubility calculated values for Sr mixture of salts at different doses

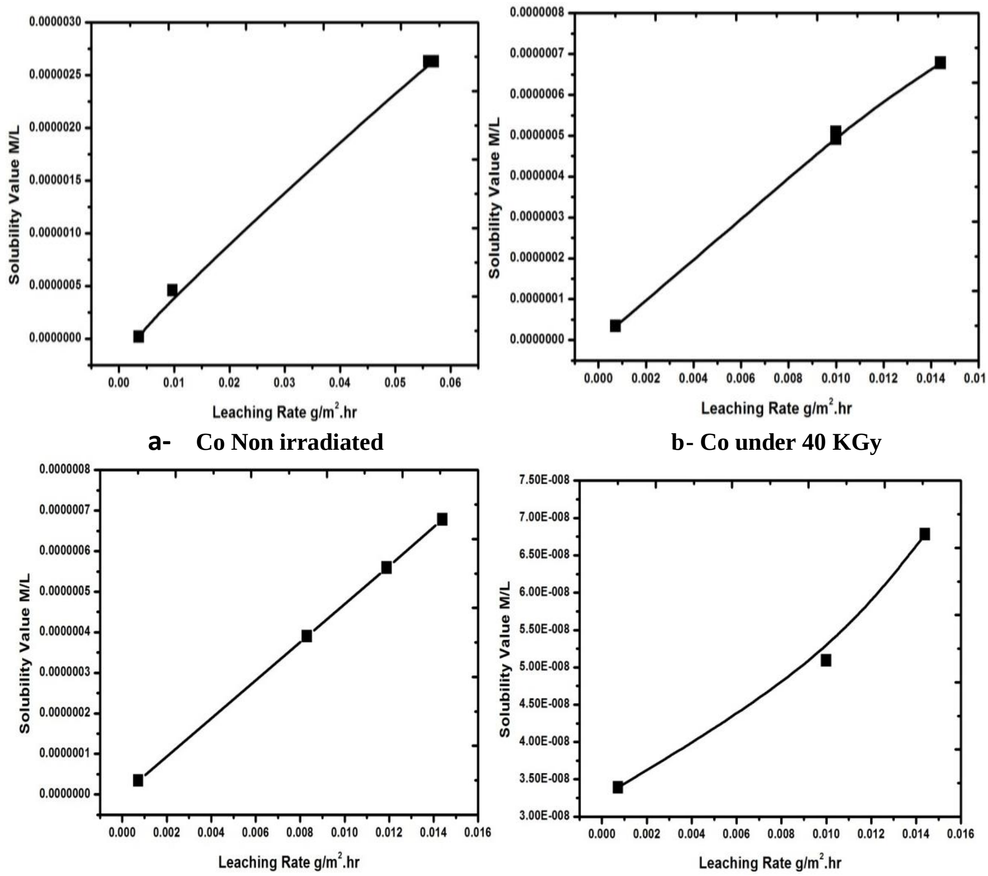


a- Co Non irradiated **b- Co under 40 KGy**



c- Co under 120 KGy **d- Co under 240 KGy**

Fig. (11) The relation between the leaching rate and solubility calculated values for Co single salt at different doses



a- Co Non irradiated b- Co under 40 KGy

c- Co under 120 KGy d- Co under 240 KGy

Fig. (12) The relation between the leaching rate and solubility calculated values for Co mixture of salts at different doses

Table (3): The conditions and the percentages of solubility to the leaching rate

Conditions	Trend of solubility and leaching rate
Cement +Sr None Irradiated	y= 2.13E-5x+ 4.033E-6
Cement +Sr under 40 KGy	y= 2.49 E-5 x- 4.75 E-6
Cement +Sr under 120 KGy	y= 2.46 E-5x+ 7.38 E-6
Cement +Sr under 240 KGy	y= 3.07 E-6x+ 5.20 E-6
Cement +Co None Irradiated	y= 4.688 E-5x+ 2.54 E-11
Cement +Co under 40 KGy	y= 4.76 E-5 x -7.52 E-10
Cement +Co under 120 KGy	y= 4.70 E-8 x- 1.04E-9
Cement +Co under 240 KGy	y= 3.67 E-4 x- 3.61 E-6
Cement +Sr in Mixture None Irradiated	y= 3.11E-5 x1.67E-7
Cement +Sr in Mixture under 40 KGy	y= 8.35 E-6x- 8.43 E-6
Cement +Sr in Mixture under 120 KGy	y= 1.14 E-4 x- 2.50 E-6
Cement +Sr in Mixture under 240 KGy	y= -5.82 E-5x+1.36E-5
Cement +Co in Mixture None Irradiated	y= 3.11E-5 x1.67E-7
Cement +Co in Mixture under 40 KGy	y= 4.78 E-5x-4.78 E-9
Cement +Co in Mixture under 120 KGy	y=4.70 E-5 x- 4.95 E-10
Cement +Co in Mixture under 240 KGy	y= 2.47 E-6x+3.06 E-8

Conclusions

The present work investigates the effect of gamma rays on the leachate pH values and the relation between the leaching rate and the solubility calculated value.

The chemical analysis of the most important elements that may be released from the cement blocks in both cases (non-irradiated and irradiated) illustrates that Ca release increases with radiation, but Al and Na release decreases under radiation.

The effect of gamma radiation on leachate pH from cement blocks simulated was performed under 40, 120, and 240 KGy, and without radiation. All the cement blocks resulted in a significant decrease in the pH values due to the radiolysis-produced H_2. The non-irradiated cement blocks resulted in an increase in the pH, with a value that can reach 14. This may be due to the release of $Ca(OH)_2$.

The Leaching rate calculation of the elements under radiation and without radiation in both cases of single and mixture of salts showed a decrease under radiation in all cases except for Co single salt.

The solubility values were calculated for elements under radiation and without irradiation in cases of single and mixture of salts. The solubility calculated values decrease under radiation for most of concentrations in single and mixture of salts, except for Co single salt. This may be due to the effect of radiation induced-blocking of the pours of cement materials.

The relation between the leaching rate and solubility calculated value is developed in the form of an equation represented by a linear line.

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