

Conditioning of Low and Intermediate Radioactive Wastes by Petroleum-Bitumen

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Abstract: The stability of the Petroleum-Bitumen toward gamma radiation before the solidification process was carried out by Thermogravimetric (TG) and Differential Scanning Calorimetric (DSC) systems at different temperatures. Immobilization of radioisotopes contained in liquid radioactive waste was investigated in the case of cementation and bituminization processes. Leaching of radioisotopes incorporated in cement and bitumen samples was studied using conductivity measurements of liquid phases.

The results showed that the bitumen contained organic solvents that need to be evaporated before mixing the radioactive wastes. The DSC/TG spectrum illustrated that the heating of the petroleum bitumen to 180 °C or/and gamma radiation before mixing the radioactive waste with bitumen did not affect the structure of bitumen specimens.

The conductivity results showed that the leaching rate was higher in the case of bitumen than the cement samples. This observation could be due to the form of binding between the radioisotope salts and bitumen or cement through the sample preparation.

Keywords: radioactive wastes treatment, conditioning of wastes, Bituminization and cementation process.

1 Introduction

Radioactive waste management plays an important role in environmental protection, because there is a large amount of radioactive waste produced by different types of nuclear reactor plants [1].

Management of low and intermediate level radioactive wastes that are produced from diverse applications of radionuclides strives to use a strategy that incorporates a number of technologies to convert wastes into suitable forms for handling, storage, transport and disposal [2].

The objectives of liquid waste processing are to immobilize the radioactive elements and to reduce the volume to be stored. The solidified product must be non-dispensable, insoluble, and with good mechanical and structural stability [3].

Treatment of low and intermediate level radioactive waste results in sludge which contains 95-98% of the radioactivity. The dewatered sludge generated contains solids concentrations depending on the management of low and intermediate level radioactive wastes for its composition and its parameters [4].

The precipitation of insoluble compounds (concentrated liquid wastes) is one of the oldest and most used processes for the treatment of radioactive aqueous wastes. Cementation has been the first and still remains the most widely used conditioning process for low and intermediate level radioactive waste [5]. Nowadays, the bituminization of radioactive wastes has been a technique used by the nuclear industry, and it has attracted interest with the cementation process in the nuclear world [6].

Among the conditioning techniques, bituminization is one that can, for some wastes, provide a form that is mechanically, physically and radiologically stable for the subsequent stages of radioactive waste management, including disposal [7].

In a previous paper [8,9], the migration of ²²²Rn, ⁹⁰Sr and ¹³⁷Cs incorporated in cement was reported. The initial distribution of ¹³⁷Cs in cement specimens was studied, and the effect of leaching on ¹³⁷Cs distribution was also studied [10].

The main objectives of the present work were to study the immobilization of ¹³⁷Cs in petroleum - bitumen specimens and to compare the leaching effect of ¹³⁷Cs in the bituminization and cementation process.

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2 Experimental Section

The radioactive element used was ^{137}Cs , supplied from radioactive liquid waste of the research laboratory, which contained (2800 Bq/L) of ^{137}Cs +inactive salts. The salinity of the liquid radioactive waste was determined by conductivity measurement using a Cyber Scan-10 instrument.

Bitumen used was obtained from the Syrian petroleum distillation station, while the cement used was a Syrian ordinary Portland Cement.

Bitumen is an organic material which consists of a mixture of mainly aliphatic and aromatic hydrocarbons of high molecular weight. The constituents appear to be enormously variable, complex, and subject to rapid change under different methods of treatment, especially by radiation. For this reason, Differential scanning calorimetry and thermogravimetry of bitumen were studied using DSC/TG-20 Mettler. Bitumen was irradiated to different gamma doses before being mixed with radioactive sludge.

The gamma irradiation of bitumen was conducted using ^{60}Co - gamma cell, Issledovatel – 10 kCi, at a dose rate of 2.8 kGy/h. The dose and dose rates were determined by means of a modified Fricke dosimeter (Fricke and Hart, 1966). Samples were irradiated at 20 °C with doses ranging from 50 to 250 kGy.

Bitumen samples were prepared by evaporating one liter of ^{137}Cs liquid (2800 Bq) to have a sludge form. This sludge was mixed with 100 g of bitumen at 180 °C for one hour in dynamic state to have a homogeneous paste. On the other hand, the cement was mixed with distilled water containing ^{137}Cs / inactive salts (water/cement ratio of about 1/3) to make a paste.

A cylindrical plastic mold of dimensions 5.5 x 4 cm (length x diameter) was used to form the samples. The samples were left to dry in air for two weeks. Triplicate samples were prepared for each form.

The samples were put individually in special pools; each contained 2 litres of distilled water under stationary conditions. The temperature of the water was set at 25°C for all samples.

Gamma rays emitted from ^{137}Cs were measured using a counting system that consisted of a NaI (TI) detector, HV supply, a rate meter, and a scaler. The efficiency of the detector was 20 %. The rate meter window was adjusted to maximize the gamma ray counts of the 662keV peak.

The conductivity of the leached water was measured with Cyber Scan-10 instrument as a function of time.

Both measurements, gamma rays and conductivity, will give a general view of the immobilization of ^{137}Cs in the samples.

3 Results and Discussion

The variation of conductivity as a function of (NaCl) salt concentration is shown in Fig. 1. The conductivity of the radioactive liquid was measured, and then the concentration of the soluble salts was found to be 7g/L. While the total salts in the liquid radioactive waste was 21g/L, measured by evaporation methods.

Thermogravimetric (TG) and differential scanning calorimetric (DSC) of bitumen before and after irradiation by gamma were illustrated in Figs. 2 and 3. The results showed that endothermic weight losses were observed at 50, 180, 400, 700 and 800 °C. The first loss can be assigned to degassing of the bitumen solvent, while the other losses are due to the decomposition of bitumen. On the other hand, the low dose of gamma irradiation did not affect any changes in the bitumen form, while the high dose let the weight losses was little.

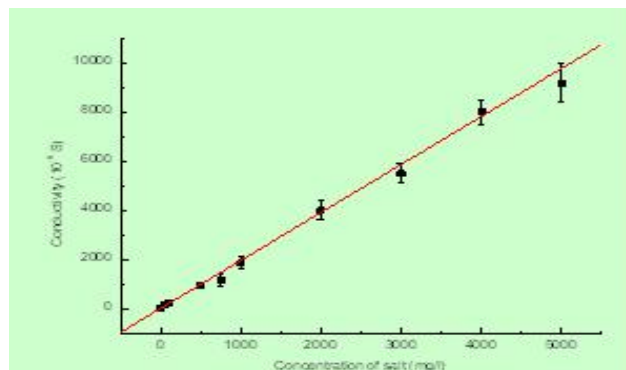


Fig.1: Variation of conductivity as a function of salt concentration.

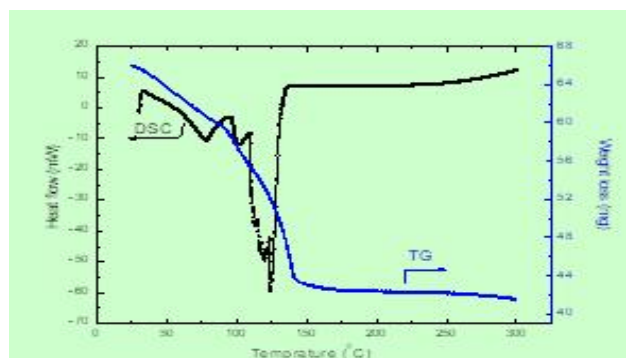


Fig.2: TG and DSC spectrum of fresh Bitumen.

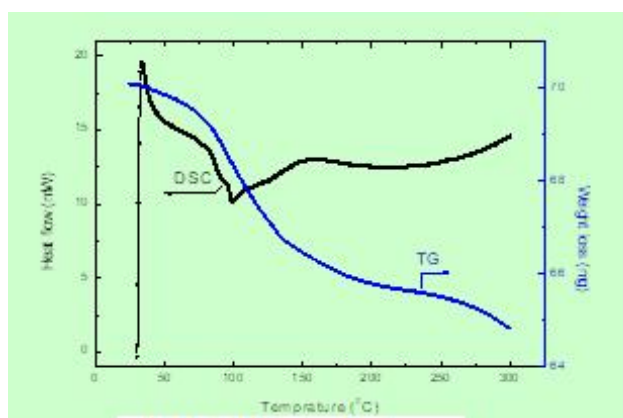


Fig.3: TG and DSC spectrum of dry Bitumen

For this reason, the bitumen sample was heated to 180 °C before being irradiated with a high dose to study the weight losses as shown in Fig. 3. The results showed that heating the bitumen to 180 °C lets the bitumen solvent to escape, and the DSC spectrum did not report a deep sharp peak. The same observation was conducted with the irradiated samples.

Figure (4) shows the thermal analysis curve of bitumen samples used in the study before and after irradiation with different doses of gamma rays within the range of 50-250 kGy. It is observed that the bitumen loses approximately 36% of its weight when heated to 180°C without irradiation or exposed to a small dose. The sample irradiated with a high dose (250 kGy) loses approximately 13% at the aforementioned temperature.

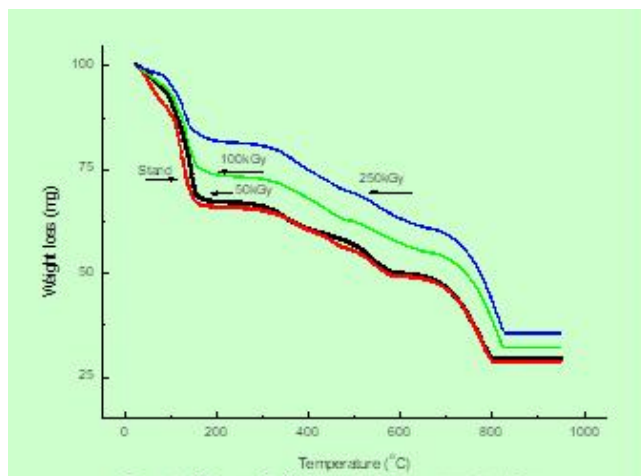


Fig.4: TG curves for Bitumen submitted to Gamma irradiation.

A sample of bitumen was heated to 180°C and another sample was irradiated to a dose of 250 kGy. The thermal decomposition curves of both samples were studied in comparison to a standard sample, as shown in Figure (5). It

was found that heating to the aforementioned temperature and irradiation to the high dose both produced the same effect on the bitumen sample.

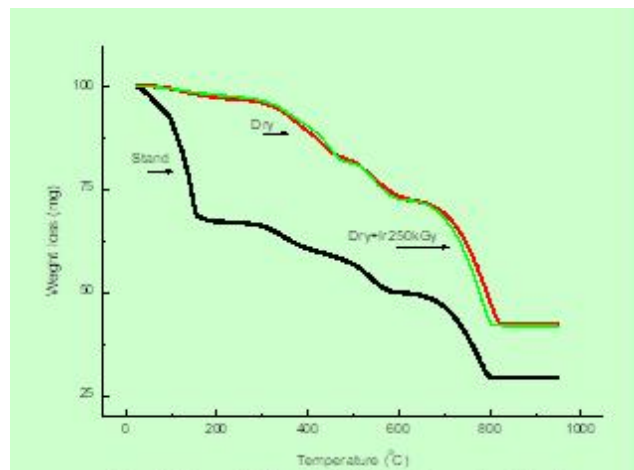


Fig.5: Comparisons TG curves of heating and Gamma irradiation Bitumen.

Figure (6) shows the thermal analysis curve of bitumen samples dried to 180°C in a dryer and then exposed to gamma rays, compared to a sample that was not dried. It is observed that heat treatment of the bitumen before irradiation makes it thermally stable up to approximately 350°C. It is also observed that the irradiated bitumen samples, when heat-treated, do not differ from the normal, non-irradiated, and heat-treated samples; that is, the irradiation process does not weaken the thermal properties of the bitumen. However, the non-heat- and non-irradiated sample, the standard sample, exhibits a change in its physical properties.

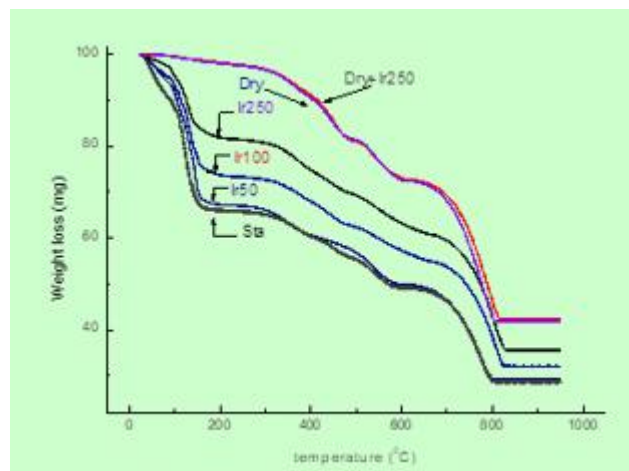


Fig.6: TG curves for Bitumen submitted to different temperature.

The leaching of fixed salts in bitumen and cement samples was measured by the electrical conductivity measurement

of the basin water, as well as that of standard basin water (without radioactive samples). Figure 7 shows the measured conductivity values for the aforementioned samples.

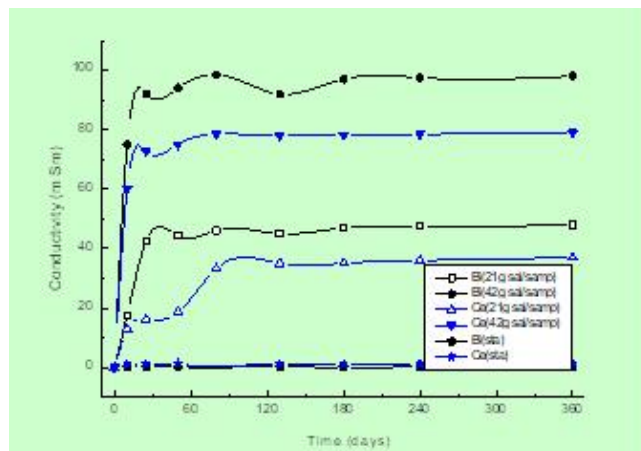


Fig.7: Conductivity of leached water for Bitumen and Cement specimens.

Analysis of Results and Discussion

1- Thermogravimetric Analysis

The thermogravimetric analysis curves provided a general overview of the physical changes that occurred in the bituminous samples exposed to heat, whether irradiated or not. The results were analyzed using a mathematical program that calculates the rate of weight loss with respect to time (phase transformation) of bitumen as a function of temperature, i.e., $\partial(\partial m)_{(T)} / \partial t$. Figures 8 and 9 illustrate these changes, showing several phases of weight loss at specific temperatures starting from 200°C. The transformation temperatures and the percentage weight loss for the studied samples are recorded in Table 1.

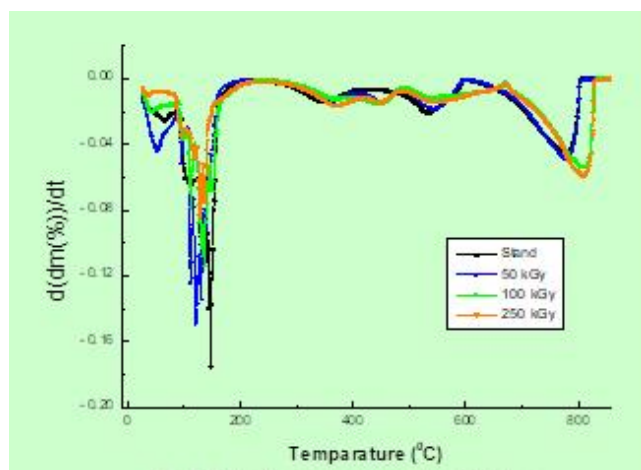


Fig.8: Rate of weight loss for irradiated Bitumen.

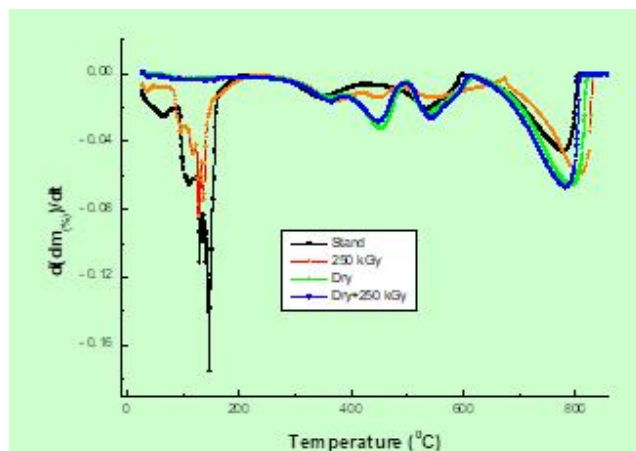


Fig.9: Rate of weight loss for irradiated and dried Bitumen.

For example, at the end of the thermal decomposition of non-thermally dried samples at 180°C, irradiation raises the final decomposition temperature to over 800°C. In other words, irradiation increases the bitumen's resistance to degradation. However, for samples dried to 180°C, irradiation reduces the overall degradation temperature by approximately 50°C. Regarding weight loss, irradiation of dried samples results in less sample loss compared to non-dried samples. This underscores the role of irradiation in maintaining the cohesion of polymeric samples, given that bitumen exhibits a similar condition.

Table 1: Transition temperature (Tm) and weight losses (Dm) for the bitumen samples.

	Stand	50 kGy	100 kGy	250 kGy	Dry	Dry+250 kGy
Tm1(°C)	150	150	150	150	150	150
Tm2	350	-	-	-	-	-
Tm3	-	380	380	380	380	380
Tm4		450	450	450	450	450
Tm5	540	-	-	-	-	-
Tm6		550	550	550	550	550
Tm7	775	-	-	-	-	-
Tm8		780				780
Tm9					800	
Tm10			810	810		
Dm1(mg)	71	67	77	84	98.4	98.5
Dm2	63					
Dm3	-	61	67.7	74.5	91.	91.5
Dm4	-	56	63	70	82	82.5
Dm5	52.6	-	-	-	-	-
Dm6	-	51.5	59.9	66.2	76.4	76.7
Dm7	34.1	-	-	-	-	-
Dm8	-	32	-	-	-	45.8
Dm9	-	-	-	-	46	
Dm10	-	-	35.83	39.8		

Finally, the breakdown of organic compounds is often complete in an inert atmosphere, as in our case. Even at high temperatures, approximately 10-20% of the original

quantity remains unbroken, and these residues mostly form tar or, to a lesser extent, highly resistant carbon bonds.

From a thermodynamic perspective, the general relationship expressing secondary and crystallographic transformations is:

$$k(T) = k_0(n)T^{(-n)} \exp\left[\frac{\Delta G(n)}{KT}\right] \quad (1)$$

Where $k(T)$ represents the phase transition rate constant, and n represents the thresholds in the thermodynamic curves, which take the following values: $n = 0, 0.5, 1$, as mentioned previously (19). ΔG (the Gibbs function) represents the free energy, and K is the Boltzmann constant. Since the free energy function relates enthalpy and entropy by the following equation, equation (1) can be written as follows:

$$k(T) = \frac{h}{KT} \exp\left(\frac{\Delta G}{RT}\right) = \frac{h}{KT} \exp\left(\frac{\Delta H}{RT}\right) \exp\left(-\frac{\Delta S}{R}\right) \quad (2)$$

Since irradiation takes place in a closed, i.e., non-existent atmosphere, according to Arrhenius' theory, relationship (2), constant velocity, can be written as the following energy relationship:

$$k = k_0 \exp\left(\frac{E}{RT}\right) \quad (3)$$

Here, E represents the activation energy (Boltzmann constant). An Arrhenius curve can be plotted by plotting $\ln(k)$ in terms of $(1/T)$ to find k_0 . However, in our case, an Arrhenius curve could not be plotted. Instead, the curve representing the relationship between weight loss and transformation temperature, as shown in Table (1), was plotted in Figure (10). Figure (10) shows a linear relationship between the amount of weight loss and the transformation temperature for all samples under different treatment conditions, whether thermal or radioactive.

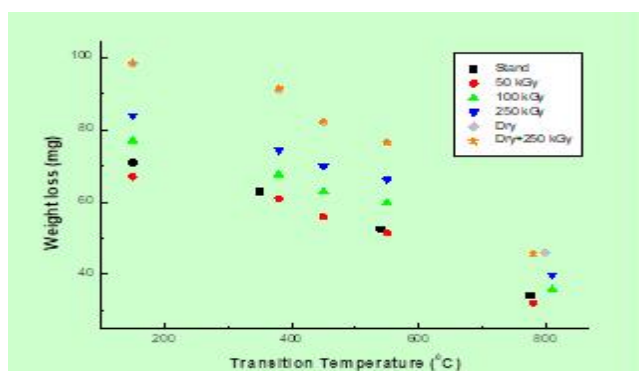


Fig.10: Transition temperature of the samples derived from Fig.8 and Fig.9.

2- Electrical Conductivity Changes and Leaching Measurements

The electrical conductivity values of the studied basin waters (in the case of bitumen, cement, and standard samples), representing leaching changes, are shown in Figure (7). It can be observed from the figure that:

- The conductivity values increase with time in both studied cases. This indicates the dissolution of some of the salts that make up the sample in both cases.
- The conductivity values recorded for bituminous samples are higher than those for cement. This is due to the formation of cement paste, which consists of mineral oxides that harden through water of crystallization, and the salts within it bind to the resulting network. Bituminous paste, on the other hand, is composed of hydrocarbons that cannot bind salts due to their chemical or network structure.
- In the case of standard samples (those without salts), the conductivity values of bitumen are slightly lower than those of cement because it primarily contains hydrocarbons, not dissolved salts, and the conductivity values remain constant.

These observations were unexpected, as bitumen is known to be a heavy organic material that prevents water penetration, or more accurately, is used to prevent water leaks. However, from a technological perspective, the samples were prepared by mixing the radioactive salts with the viscous liquid bitumen, and then allowing the samples to air dry. This method represents a physical event (mixing) without altering the structural composition.

In this case, a percentage of salts may remain on the surface of the samples exposed to the basin water. As for cement, after pouring the cement paste into plastic molds and during the drying period, physical and chemical transformations usually take place that work to bind it together through the bonding of water of crystallization. As for the process of wetting the samples, it is an assistive process that works to increase the amount of water molecules, which in turn works to increase the cohesion of the cement crystal lattice and increase the bonds between aluminum oxides, silicates, and iron, in addition to the metal ions present in the sludge (especially radioactive cesium ions), which exchange with the ions of the cement components, as mentioned previously (5).

During leaching, a considerable amount of ions dissolves in the water, which may not necessarily be cesium ions. This observation was confirmed by radiometric measurements of the basin water and solid samples after the experiments were completed. Figure (11) shows the radioactivity (CPS) values after the total experimental period of 360 days. The data in this figure show an increase in the measured CPS value in the cementitious samples compared to the

bituminous samples. This was previously confirmed in Figure (7) regarding the measured electrical conductivity of the studied samples.

Furthermore, the radioactivity of the slurry containing the bitumen samples was higher than that of the slurry containing the cement samples. This indicates that preserving radioactive waste by mixing it with cement and allowing the mixture to harden is preferable to mixing it with bitumen, which cannot retain radioactive isotopes.

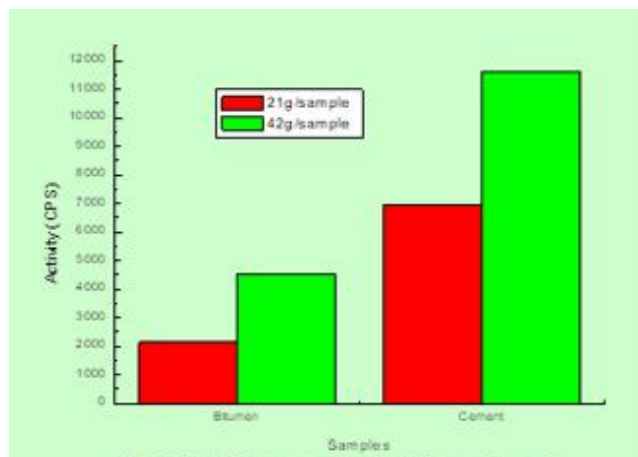


Fig.11: The samples activity after 360 days of leaching.

4 Conclusions

The results of this work showed that ionizing radiation (gamma rays) affects the physical and/or chemical properties of bitumen.

The method of preserving radioactive waste in cement is considered one of the best practical methods to date in terms of ease of handling and the preservation of the physical and chemical properties of the cementitious composition, even after mixing the cement with radioactive isotopes.

Bitumen is unable to retain radioactive isotopes and salts associated with radioactive waste liquid compared to cement. This observation may be attributed to the properties of cement, which may bind radioactive isotopes (positive ions) to the ions present in its structural composition, while bitumen traps radioactive isotopes and positive ions (possibly spatially) within its spatial structure, which makes it easy for these ions to decompose and drift under any influence, however weak.

Acknowledgement

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