

Development of a novel relationship between salt content and radium concentration in crude oil

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Radium is one of the most important natural radioactive sources associated with crude oil and its extraction processes. The importance of this radioactive element is highlighted by its ability to precipitate inside oil pipelines, forming what is known as scales. Depending on its concentrations in the extracted oils, its concentrations vary according to the type of oil reservoir and its geological formations. The aim of the current study is to find a mathematical relationship linking the salt content of crude oil with the concentration of radium. Thirty samples of crude oil were taken from the fields in Thi Qar province, Iraq, and tested to determine the concentrations of radium resulting from the ^{238}U and ^{232}Th series using gamma ray spectroscopy based on a sodium iodide scintillation detector with dimensions of 3×3 inch. At the same time, the salt content was also determined in each of the studied samples. The obtained results were used to create a mathematical relationship linking the radioactive concentration of radium isotopes to the salt content in crude oil. The relationship showed an accuracy of nearly 90% and demonstrated a direct relationship between the salt content and the concentrations of radium isotopes.

Keywords: Salinity; radium-salinity relationship; radioactive waste management; radioactive concentration; salt content.

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1. Introduction

Crude oil is a strategic natural resource that is formed through complex geological processes, and often contains varying amounts of Naturally Occurring Radioactive Materials (NORM) [1]. Among these radioactive materials, ^{226}Ra and ^{228}Ra are the main isotopes that arise as a result of the decay of ^{238}U and ^{232}Th chains present in oil-bearing rocks [2,3]. These radioisotopes accumulate in groundwater fluids and crude oil through mechanisms that include dissolution in groundwater and interaction with surrounding minerals, making the study of their distribution in crude oil an issue with environmental and industrial dimensions [4,5]. On the other hand, salinity is an important physical and chemical property in petroleum systems, as it expresses the concentration of ions dissolved in the fluids associated with oil. Salinity directly affects the chemical properties of crude oil, and may contribute to changing the dynamics of the presence of naturally occurring radioactive elements such as radium [6,7]. Scientific hypotheses indicate the possibility of a relationship between salinity and the concentration of natural radioactivity, as it is believed that high salinity may enhance the solubility of radioactive elements in liquids, or on the contrary, may lead to their precipitation as a result of chemical reactions with the existing ions [8]. Despite the numerous studies that have addressed natural radioactivity in crude oil and associated water, the exact relationship between radium concentration and salt content has not been adequately studied [9]. In some studies, the role of salinity has been indicated as a catalyst for increasing radium concentration as a result of enhancing the movement of radioactive isotopes in the liquid environment. While in other studies, it has been noted that high salinity may lead to the precipitation of these el-

ements as a result of changes in the chemical equilibrium [10,11]. The chemical processes explaining the increase in radium solubility with salinity include the fact that radium (Ra^{2+}) is part of the alkaline earth metal group, and its behavior in saline solutions resembles that of barium (Ba^{2+}) and strontium (Sr^{2+}), as its solubility rises with increasing salinity due to several key chemical mechanisms, such as ion exchange/absorption, the influence of common ions on sulfates, and the formation of radium chloride complexes with Cl^- . Kraemer and Reid [12] demonstrated that radium solubility increases with salinity due to ion exchange in sediments, highlighting the role of sodium in radium displacement. Hanor [13] indicated that the solubility of radium is mainly governed by the equilibrium between radium and barium sulfate. Additionally, he demonstrated that the formation of a $(\text{Ra}, \text{Ba})\text{SO}_4$ solid solution accounts for the increased release of radium in saline waters. Chloride complexes play a minor yet significant role in hypersaline marine waters, particularly in regions with fluctuating temperatures and pressures, as shown by Hedström H. [14] and Kitamura A. [15]. The solubility of substances increases with salinity, primarily due to the formation of solid solutions that inhibit the precipitation of pure RaSO_4 . Additionally, the formation of chloride complexes occurs at high Cl^- concentrations. The significance of these mechanisms varies based on sulfate and barium concentrations, sediment quality, and salinity. This study aims to determine the relationship between radium concentration and salt content in crude oil through a precise experimental study that includes measuring radium concentration using gamma spectrometry and analyzing salinity using advanced techniques [16,17]. The study also seeks to develop a new mathematical relationship that explains the behavior of radium under salinity changes, which contributes to improv-

ing our understanding of chemical and physical reactions in oil reservoirs. The results will contribute to enhancing scientific knowledge about this phenomenon, as well as providing practical applications to improve radioactive waste management and reduce environmental risks associated with the oil industry.

2. Material and methods

Thirty crude oil samples were collected and prepared from crude oil fields in Thi Qar province. Ten samples were taken from each field in sealed containers to study the relationship between radium concentration and salt content. The samples were left for one month to achieve radioactive equilibrium between radium isotopes and its radioactive products spatially, Actinium-228 (energy line 911 keV) and Bismuth-214 (energy line 609.3 keV).

2.1. Radium measurement

Every sample was approximately weighed to fill a one-liter container (Marielle Becker). Using a “3 × 3” inch Teledyne isotope NaI (Tl) gamma-ray spectroscopy system with a 7.5 percent resolution at 661.76 keV and gamma line for a ¹³⁷Cs source [18,19]. The samples were measured and analyzed to obtain the radium isotopes concentrations. In the current study, four methods were used to calibrate the measurement system to obtain the highest possible accuracy, such as calibrating the energy to its location (channel), the detector resolution, the experimental efficiency, and the minimum detectable activity. The minimum detectable activity was measured for ²²⁶Ra at energy line 609.3 keV and it was 0.112 Bq, and for ²²⁸Ra at energy line 911.2 keV, and it was 0.114 Bq. The concentration of ²²⁶Ra and ²²⁸Ra in crude oil was calculated based on the following equation [20].

$$A_{Ra} = \frac{N}{t \cdot I_{\gamma}(E_{\gamma}) \cdot \epsilon(E_{\gamma}) \cdot m} \times DCO, \quad (1)$$

where A_{Ra} (Bq/L) is the activity concentration, (N) refers to the net area under the gamma-ray peak, (t) represents the measurement time, $I_{\gamma}(E_{\gamma})$ is the intensity of the targeted gamma-ray energy, $\epsilon(E_{\gamma})$ refers to the efficiency of the energy of the gamma-ray line, and (m) represents the mass of the sample. DCO is the density of crude oil under investigation.

2.2. Salt content measurement

Salt content measurement technique uses a glass system made by Petrotest that is based on the standard IP 77 to separate the salt content of the crude oil [21]. This extraction process involves heating the sample in a glass flask with a volume of 55 cm³ using an electric heater. The flask is filled with roughly 155 milliliters of crude oil and 155 milliliters of distilled water. Following mixing, 20 milliliters of acetone and 100 milligrams of toluene were added. The electric

heater heats the flask to the mixture’s boiling point, and a vertical condenser placed on top of the flask cools and returns the vapors that are produced. At least forty-five minutes are spent on this procedure. To ensure that a longer period is needed to complete the salt extraction. Following the extraction step, the water phase is separated, and the chlorine ion content is neutralized using a Mohr volumetric titration method. The contents of the cooled water phases and organics are then separated and released through the valve inserted beneath the flask. In this manner, chloride ions are titrated using regular 0.01 M silver nitrate with a few drops of potassium dichromate solution at a 50 g concentration [22,23]. The surrounding tint shifts from yellow to light crimson once the chlorine ions have been neutralized. Equation (2) gets the sodium chloride concentration in milligrams per cubic meter.

$$X = \left(\frac{V_A}{V_1} - \frac{V_B}{V_2} \right) / V_S \times N \times 58500 \times V_E, \quad (2)$$

where V_E is the total volume of the extract, V_S is the volume of the crude oil sample used, N is the normality of the silver nitrate solution, V_A is the volume of silver nitrate solution used for the extraction of the titration in milliliters, V_B is the volume of silver nitrate used as the witness of the titration, V_1 is the volume of extraction consumed used in the titration, and V_2 is the volume used.

3. Result and discussion

In the current study, the analysis of the crude oil samples that were taken from the oil fields, showed a clear relationship between the activity concentration of Radium and the salt content in crude oil. Table I illustrates the results of the activity concentrations of (²²⁶Ra and ²²⁸Ra) and the salt content in oil fields under investigations. It was noted that the highest salt content leads to the highest activity concentrations of radium. Figures 1 and 2 indicated the relation between the activity concentrations of ²²⁶Ra and ²²⁸Ra and the salinity in the crude oil with an accuracy more than 90%. The results of the statistical analysis showed that the relationship between

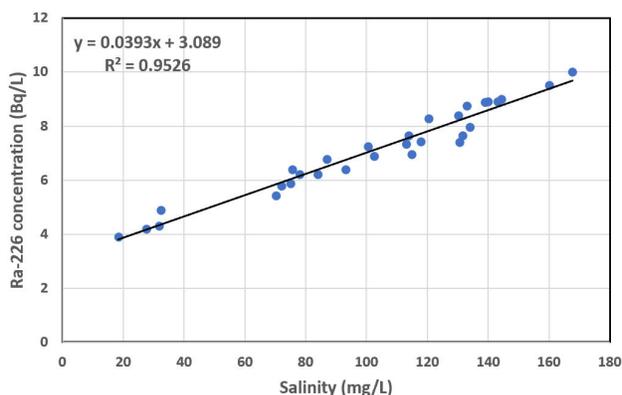


FIGURE 1. The relationship between the salt content and ²²⁶Ra concentration in crude oil.

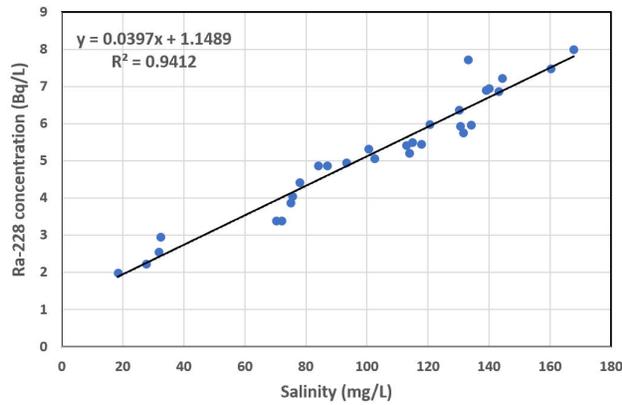


FIGURE 2. The relationship between the salt content and 228Ra concentration in crude oil.

TABLE I. Activity concentrations of (^{226}Ra and ^{228}Ra) and the salinity of the targeted crude oil samples.

Sample ID	Salinity (mg/L)	^{226}Ra (Bq/L)	^{228}Ra (Bq/L)
S1	32.4 ± 0.176	4.9 ± 0.452	2.96 ± 0.581
S2	27.56 ± 0.190	4.2 ± 0.488	2.24 ± 0.668
S3	31.82 ± 0.177	4.32 ± 0.481	2.56 ± 0.625
S4	102.4 ± 0.099	6.9 ± 0.381	5.07 ± 0.444
S5	75.56 ± 0.115	6.39 ± 0.395	4.06 ± 0.496
S6	113.82 ± 0.094	7.65 ± 0.362	5.22 ± 0.438
S7	117.76 ± 0.092	7.42 ± 0.367	5.46 ± 0.428
S8	18.39 ± 0.233	3.9 ± 0.506	1.99 ± 0.709
S9	131.53 ± 0.087	7.65 ± 0.362	5.76 ± 0.417
S10	93.15 ± 0.105	6.4 ± 0.395	4.95 ± 0.449
S11	77.94 ± 0.104	6.21 ± 0.395	4.43 ± 0.475
S12	120.47 ± 0.113	8.29 ± 0.401	5.99 ± 0.409
S13	167.7 ± 0.077	10.01 ± 0.347	8.00 ± 0.354
S14	71.98 ± 0.077	5.79 ± 0.316	3.39 ± 0.543
S15	83.94 ± 0.118	6.22 ± 0.416	4.87 ± 0.453
S16	86.87 ± 0.109	6.78 ± 0.401	4.88 ± 0.453
S17	70.28 ± 0.107	5.44 ± 0.384	3.39 ± 0.543
S18	100.43 ± 0.119	7.26 ± 0.429	5.32 ± 0.434
S19	112.96 ± 0.100	7.34 ± 0.371	5.42 ± 0.430
S20	133.00 ± 0.094	8.76 ± 0.369	7.73 ± 0.360
S21	130.24 ± 0.087	8.39 ± 0.338	6.38 ± 0.396
S22	138.91 ± 0.088	8.88 ± 0.345	6.91 ± 0.380
S23	144.38 ± 0.085	9.01 ± 0.336	7.22 ± 0.372
S24	143.14 ± 0.083	8.91 ± 0.335	6.87 ± 0.382
S25	139.97 ± 0.084	8.91 ± 0.335	6.95 ± 0.379
S26	134.00 ± 0.085	7.96 ± 0.354	5.97 ± 0.409
S27	74.95 ± 0.086	5.89 ± 0.412	3.87 ± 0.508
S28	160.12 ± 0.079	9.51 ± 0.324	7.49 ± 0.365
S29	130.63 ± 0.087	7.41 ± 0.367	5.94 ± 0.410
S30	114.75 ± 0.093	6.96 ± 0.379	5.51 ± 0.426

crude oil salinity and ^{226}Ra concentration is strongly linear ($R^2 = 0.95$). The slope of the line was 0.039 Bq/L for each 1 mg/L increase in salinity, with a 95% confidence interval ranging from 0.036 to 0.043. The intercept was 3.09 Bq/L, with a confidence interval between 2.73 and 3.45. This analysis confirms the ability of the proposed equation to accurately predict radium concentration when the salt content is known. Statistical analysis showed that the relationship between crude oil salinity and radium-228 concentration was strongly linear ($R^2 = 0.94$). The slope of the line was 0.040 Bq/L per 1 mg/L salinity, with a 95% confidence interval of 0.036-0.043. The intercept was 1.15 Bq/L, with a confidence interval of 0.75-1.55. These results support the accuracy of the proposed equation in predicting radium-228 concentration when salinity is known.

The results also indicated that the crude oil fields with high salinity showed higher activity concentrations of Ra isotopes than the oil fields with low salinity. This correlation is attributed to the increased dissolution of radium elements in the water remaining in the crude oil, as radium ions are closely associated with chloride and sodium ions present in brines. Also, the results showed that the solubility of radium in a saline environment is a major factor in its high concentrations, which means that crude oil extracted from fields with high salinity contains higher levels of naturally occurring radioactivity (NORM). This phenomenon emphasizes the importance of studying salt content when evaluating radium isotopes levels in crude oil. According to the results, new mathematical equations were developed to connect the radium isotopes concentration to the salt content in crude oil, which is an important step that can be used to predict radium levels based on salinity values in crude oil. It was discovered that there is a direct relationship between them, indicating that the concentration of radium increases with the concentration of salt content. This proposed linear relationship contributes to the development of rapid and effective methodologies for monitoring natural radioactivity of radium isotopes in crude oil, which helps improve environmental monitoring processes and reduce the risks associated with radioactivity in the crude oil.

$$A(^{226}\text{Ra}) = 0.0392 + 3.088 \times \frac{\left(\frac{V_A}{V_1} - \frac{V_B}{V_2}\right)}{V_S} \times N \times 58500 \times V_E, \quad (3)$$

$$A(^{228}\text{Ra}) = 0.0397 + 1.1488 \times \frac{\left(\frac{V_A}{V_1} - \frac{V_B}{V_2}\right)}{V_S} \times N \times 58500 \times V_E. \quad (4)$$

4. Conclusion

The present study showed that a clear linear relationship between the salt content of crude oil and the concentration of ^{226}Ra and ^{228}Ra isotopes. The results indicated that the

crude oil with high salt content contains higher concentrations of radium isotopes compared to other crude oils with low salinity. This is attributed to the ability of radium ions to dissolve in the brine solutions accompanying crude oil, where they interact with chloride and sodium ions. Based on these results, an innovative mathematical equation was developed that links salt content to radium concentration, which enables the prediction of radioactivity levels in crude oil based on salinity. This discovery is of great importance in the fields of

natural radioactivity monitoring (NORM) in the petroleum industry, and contributes to improving environmental procedures and reducing the risks associated with handling highly saline crude oil.

Conflict of interest statement

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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