Determination of the Radon Concentration of Gas 222Rn Emitted From Sludge Samples Located in the Radioactive Waste Collection Warehouse in Khader Al-Maa South of Basra City – Iraq

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Abstract

Thirty-one samples of sludge were taken, stored at the natural radioactive waste collection site, NORM, in Khidir Al-Maa, south of Basra city, resulting from the operations of extracting and filtering crude oil in oil fields in Basra Oil Company, to determine gas concentrations Radon 222Rn and other radioisotopes in Sludge models. The rapid electronic technology was adopted through the RAD7 device, the active method, and the gamma analysis technique using HPGe High Purity Germanium Detectors to determine the specific activity of radioactive elements. The measurements for this study showed 98463±7031 Bq.m⁻³ in sample no. S10 a barrel of sludge was brought from the third Degassing Station plant in the northern Rumaila oil field and least concentrated is 8317±594 Bq.m⁻³ in sample no. S14 sludge belonging to the sixth Degassing Station / West Qurna-1, the study also revealed the presence of high concentrations of the specific effectiveness of radioactive isotopes (214Pb, 212Bi, 226Ra) and (212Pb, 212Bi, 228Ac) of the uranium-238 series and Thorium-232 series in a sludge form selected from a barrel returning to the third gas isolation plant - North Rumaila Oilfield and stockpile at the collection site, where the specific activity value of radium was 226 Ra 133851 Bq.k⁻¹ of the uranium-238 series and for lead 212Pb 56432 Bq.k⁻¹ of For uranium-238 series compared to their concentrations, 32 Bq.k⁻¹ 45 Bq.k⁻¹ in a natural soil model. The effective dose to which on-site workers would be exposed in contact was calculated and found to be 196.92 Svh⁻¹, a dose that exceeded the proposed 50 Sv. h⁻¹ dose limits by the US EPA. The probability of developing cancer due to exposure to radon per million people was found and it was found to be a high value compared to the suggested value (170-230) per million people previously by ICRP.

Keywords: Sludge, Rn, Radium, RAD7, HPGe High Purity Germanium Detectors, Effective dose.
تحديد تركيز غاز الرادون 222Rn المنبعث من نماذج الحماة الموجودة في مخزن تجميع النفاضات المشعة في خضر الماء جنوب مدينة البصرة-العراق

الخلاصة:

جمع في هذه الدراسة (31) عينة من الحماة الطبيعية (Sludge) بالمخزون في موقع تجميع النفاضات المشعة وتعتبر من أهم النفاضات المشعة الأخرى كأذراع 222Rn ونظام مشعة أخرى كالإكتنيوم 228Ac والإكتنيوم 226Ra المتطرف الكترونياً يدعى RAD7 وتقنية تحليل طيف الطيف كفاءة استخدامها كأذراع جرمانيوم عالية الفرقة. الدراسة تبين نتائج الدراسة هذه إن أكبر تركيز لغاز الرادون في الحماة برميل حماة جلب من محطة عزل غاز الثالثة (Sludge) في عينة رقم 101 94863±7031 Bq.m 3 في عينة رقم 141 8317±594 Bq.m 3. عزل الغاز السادسة / حقل غرب الفرقة-1، كما استخدمت الدراسة وجود تركيز مرتفع للإلكترون D جرامانيوم النوعي للحصول على نماذج 228Ac، 212Bi، 212Pb، وحل المريلة الشمالي النفطي وفقاً للتركيزات. النموذج 222Rn في نماذج البداية من برميل عادي إلى محطة عزل غاز الثالثة - حقل المريلة الشمالي 133851 Bq.k 1.1 لسلسة اليورانيوم - 50 Bq.k 1.1 للرصاصات 212Pb بتركيزهما 196.92 μSv.h 1 وحدة الإشعاع الكلي للمنشآت في الولايات المتحدة. تم حساب الجرعة المغذية في الموقع في حالة التماس مع الحماة ووجد أنها 5 μSv. h 1 للجرعة المغذية. المواد المستنفدة في مواد إسقاط التعرض لغاز الرادون لمليون شخص ووجد أنها عالية مقارنة بالحدود المحددة (230-170).

1. Introduction:

The oil and gas industry is one of the most important and most concentrated industries for natural radioactive materials (NORM), as its workers are exposed to the danger of (NORM) [1] which appear remarkably as radioactive waste accompanying the extraction and refinement of crude oil. These radioactive wastes exist in various forms, such as sludge and scale deposits in pipes and valves, as well as faces accompanying the water associated with crude oil separation production processes, and generally contain quantities of palpable concentrations, especially of radium isotopes Ra, that may cause the environment to be exposed to pollution and then radiation exposure to workers who, they work near these contaminated sites. NORM is present in oil and gas formation basins, like other mineral elements, with varying concentrations. These radioactive materials come out during the search for crude oil from deep underground depths with production fluids. These materials are...
deposited in oil production lines and points within a single production line, and usually. The highest effectiveness of $^{226}$Ra is in the network of oil exit valves from the well and its branches to other oil equipment, and its concentration increases as scaly deposits in oil transportation pipelines, especially at the sites of bends and protrusions, and it is deposited very heavily in the crude oil insulators. The appearance of these substances in squamous sediments and sludge naturally leads to an increase in radioactive levels due to the accumulation of these substances in them [2-5] Oil and the general population come from radon gas of natural origin, which is a colorless, tasteless, and odorless gas that is seven and a half times heavier than air. Obstruction, which makes the level of radiation exposure high for large groups of the public. The inhalation of radon gas or contact with it is a certain risk, because this gas is considered the largest contributor to the exposure of the general population to natural radiation, and this gas may contribute in the range of 50-55% of the total dose that a person may be exposed to from all natural sources of radiation [6-8] Exposure to this gas causes a serious health and environmental problem, and many studies have proven a close relationship between exposure to radon gas emitting alpha particles and lung cancer, that the process of inhalation and the entry of air saturated with radon gas into a person’s lungs leads to precipitation to deposition. A large amount of it is on the walls and lining of the respiratory system and thus leads to the absorption of large quantities of doses by the bronchi [9] two measurement methods were used in this study called the first direct method through a fast advanced device called RAD7, and the second using a high purity germanium detector HPGe High Purity Germanium Detectors. The aim of this research is to determine the concentration of radon gas and other radioactive isotopes such as radium $^{226}$Ra and $^{228}$A, in samples of sludge stored at the radioactive waste collection site in Khader Al-Maa - south of Basra city.

2. Study station

The study station is located in the radioactive waste collection site - Khader al-Maa - Basra city - Iraq, Figure (1) near the Kuwaiti border and 17 km away from the center of Basra city. The site is one of the largest temporary storage site for natural
radioactive waste in the city of Basra and Iraq in general. It covers an area of 250 x 120 square meters, containing more than 25,000 barrels of slag contaminated with radioactive materials of natural origin. The capacity of one barrel is 220 liters, as well as hundreds of thousands of tons of solid radioactive waste. The site is surrounded by concrete blocks.

![Map of Basra Governorate showing the study areas.](image1)

Fig. (1): Map of Basra Governorate showing the study areas.

3. **Experimental work and materials used**

Sludge samples were taken from the radioactive waste collection and storage site - Khader al-Maa in Basra city - Iraq in April 2021. When collecting these samples, they were subjected to drying at a temperature of 110°C for twenty four hours by using a thermal oven, and sieved with a 2µm sieve to filter them from impurities and foreign bodies, then ground into a very fine powder using a grinder, and samples were prepared with scientifically approved specifications for the purpose of determining the concentration of radon and other radioactive isotopes. Figure (2).

![The Sludge in the radioactive waste collection store - Khaddar alma in the study area.](image2)

Fig. (2): The Sludge in the radioactive waste collection store - Khaddar alma in the study area.
3.1 Advanced radon Technology

RAD7 technology, which is a fast electronic device that is adopted to monitor radon in air, soil and water Figure (3). The work of this technique is based on the RAD7 device, which records the measurement of radon gas concentrations emitted from the samples in this study in real time. Where the device withdraws a quantity of the generated gas Grab from above the sample and sends it to the LOCAS cell to determine the concentrations of radon gas in the ground slag models, after making sure that there is no water vapor associated with the gases, and the process of pumping the device continues for five minutes after which the device starts for five another minutes. This counting process represents a determination of an elemental concentration of $^{218}\text{Po}$ (3.05 min), $^{212}\text{Po}$ (164 μs), so the measurement process is after (540-900) seconds, and this device is characterized by its ability to determine the energy of particle alpha electronically, which can distinguish between isotopes of elemental radon Polonium-$^{218}\text{Po}$-Polonium-$^{214}\text{Po}$-between radon $^{222}\text{Rn}$ and oxon $^{220}\text{Rn}$, the RAD7 detector must be dried (purging) with new air for 600 seconds by connecting the drying unit in a closed loop with the RAD7 device. Desiccant, when the humidity level appears to us less than 6% in this case we start the test, the pump works for 300 seconds during which the radon is extracted from the sample and it is sent to the measuring room in RAD7 and then the RAD7 device stops for a period of more than 300 seconds in order to reach the state of equilibrium and then The process is repeated for four cycles, at a rate of 300 seconds per cycle, thus the total test duration reaches 1800 seconds, and at the end of each operation, RAD7 provides us with the required information and data.

![Fig. (3): The shape of the RAD7 device used in the search.](image-url)
3.2 Gamma spectrum analysis technique using HPGe high-purity germanium detector.

The technique of gamma spectrum analysis is adopted by using the high-purity germanium detector CANBERRA connected to a multi-channel spectral analyzer and an IBM calculator for the purpose of recording gamma-ray spectra and results, and connected to a liquid nitrogen flask for the purpose of cooling to a low temperature, which is a low temperature that is adopted to maintain the crystallization of the detector during work. This technique is an important and widely used technique in the detection of isotopes that emit gamma rays. Standard sources, type $^{60}$Co, possessing two gamma lines (1332, 170) Kev and cesium $^{137}$Cs at power 662 Kev were used for the purpose of obtaining the amount of energy per channel and in order to calculate the real area under the peak, 1kg of sludge from each model was placed in the (Marineli Beaker), which is a plastic cup designed in a manner that makes the used model close to the effective detector area in order to give a high counting efficiency of gamma rays emitted from the radioactive nuclei in the sample. The natural background radiation is subtracted from the area of all the samples under study to obtain the net area under the peak.

4. Results and Discussions

The outputs of the results were included in Tables (1) and (2) where Table (1) shows radon concentrations in samples of sludge at the radioactive waste collection site - Khader Al-Maa, south of Basra city, where the table shows the minimum concentration of radon $8317 \pm 594$ Bq.m$^{-3}$ in the sample of sludge transferred from the sixth degassing station, West Qurna, and that the maximum value of radon concentration was $98463 \pm 7031 \pm$ Bq.m$^{-3}$ in the sludge sample belonging to the waste of the third degassing plant from the northern Rumaila oil field in the assembly store, which is sample No. S10, Figure (4),shows the Relationship note between the concentration of radon gas emitted from sludge samples in study area at the NORM radioactive waste collection site in the city of Basra - southern Iraq, measured by RAD7 technology, as for the concentrations of other radioactive isotopes such as radium-226 in slag samples in The location of the study area Table.
(2) We note that there is a very clear increase, as we have very high concentrations of radium-226, reaching 133,851Bq.K$^{-1}$, which is one of the radionuclides in the decay series of uranium-238 due to its environmental behavior and its long half-life. He is 1620 years old and his health risk lies in his deposition in bones [10] they were higher than the suggested global average of 32Bq.kg$^{-1}$, and also recorded high specific efficacy values for $^{212}$Pb above the suggested limits of 45Bq.kg$^{-1}$ by [11] UNSCEAR, 1993. It is likely that the reason for this increase in gas concentrations Radon $^{222}$Rn and radioisotopes ($^{214}$Pb, $^{214}$Bi, $^{226}$Ra) belonging to the uranium-238 series and radioisotopes ($^{212}$Pb, $^{212}$Bi, $^{228}$Ac) belonging to the thorium - 232 in sludge samples selected from the radioactive waste collection site - Khidr Alma, we also showed an increase in the concentration of other radionuclides compared to their concentration in sludge with the results of previous international studies, Table No. 3 [12].

U-238 and Th-232 in the sludge models selected from the radioactive waste collection site - green water in the study area, since the sludge present at the site resulted from production and maintenance operations accompanying the work in the crude oil production and separation plants, Basra Oil Company - southern Iraq, and that this is a natural product because oil is brought from the depths of the earth, and contains quantities of natural radioactive materials NORM, and the reasons for the emergence of varying concentration from one model to another we suggest that to the nature of the geological formation of the fields from which the models were brought. The effective dose was calculated upon contact with the slag, considering that 1Bq.m$^{-3}$ is equivalent to 0.025mSv y$^{-1}$ according to [13-14] EPA, CEC, meaning that 1Bq/m$^3$=2.854x10-3µSv h$^{-1}$, and from that we conclude that the greatest concentration of radon is 98463+7031±Bq. m$^{-3}$ in the sludge samples under study is equivalent to an effective dose of 196.92 µSvh$^{-1}$ on contact, which is a dose higher than the permissible dose level of 50 µSv h$^{-1}$ [15] recommended by the EPA in the United States. Figure (6) The relationship between gas concentration Radon and effective dose of sludge samples at the collection site.
The probability of developing cancer resulting from exposure to radon gas was estimated per million individuals by multiplying the annual effective dose by mS.y (18×10^-6). The probability of infection recorded in this research, when adopting the maximum dose of radon concentration 196.92µSvh^-1 results in a high value for each million people, which is considered a very dangerous value based on the proposed limits, which are (170-230) per million people [16-18].

Table (1) Rn -222 concentration (Bq/m³) in Sludge sample barrels of radioactive waste, radioactive waste collection site - khadir Alma in the study area.

<table>
<thead>
<tr>
<th>Sample No</th>
<th>Name of on-site stock Sludge barrel model</th>
<th>Radon concentration (222Rn) in Bq/m³</th>
<th>Annual effective dose µSv/h</th>
<th>Lung Cancer x10^-4</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>Central Degassing Station South</td>
<td>21314±152</td>
<td>42.62</td>
<td>774.21</td>
</tr>
<tr>
<td>S2</td>
<td>Sothern Degassing Station</td>
<td>23760±1697</td>
<td>47.52</td>
<td>863.22</td>
</tr>
<tr>
<td>S3</td>
<td>Qurenit -1Degassing Station</td>
<td>30412±2172</td>
<td>60.82</td>
<td>1104.82</td>
</tr>
<tr>
<td>S4</td>
<td>Shamei-1 Degassing Station</td>
<td>29543±2110</td>
<td>59.08</td>
<td>1073.21</td>
</tr>
<tr>
<td>S5</td>
<td>Ratka Degassing Station</td>
<td>31327±2237</td>
<td>62.65</td>
<td>1133.89</td>
</tr>
<tr>
<td>S6</td>
<td>Qurenit-2 Corporation station</td>
<td>33713±2408</td>
<td>67.42</td>
<td>1224.71</td>
</tr>
<tr>
<td>S7</td>
<td>Shimea-2 Degassing station</td>
<td>36821±2630</td>
<td>73.64</td>
<td>1337.70</td>
</tr>
<tr>
<td>S8</td>
<td>Central Degassing Station North</td>
<td>28765±2054</td>
<td>57.53</td>
<td>1045.06</td>
</tr>
<tr>
<td>S9</td>
<td>Second Degassing Station</td>
<td>29433±2102</td>
<td>58.86</td>
<td>1069.22</td>
</tr>
<tr>
<td>S10</td>
<td>Thread Degassing Station North Rumaila</td>
<td>98463±7031</td>
<td>196.92</td>
<td>3544.63</td>
</tr>
<tr>
<td>S11</td>
<td>Fourth Degassing Station North Rumaila</td>
<td>31162±2225</td>
<td>61.75</td>
<td>1121.72</td>
</tr>
<tr>
<td>S12</td>
<td>Fifeth Degassing Station North Rumaila</td>
<td>44572±3183</td>
<td>88.33</td>
<td>1604.56</td>
</tr>
<tr>
<td>S13</td>
<td>Central Pump Station</td>
<td>38120±2722</td>
<td>75.54</td>
<td>1372.22</td>
</tr>
<tr>
<td>S14</td>
<td>Sexes Degassing station of East Qurna</td>
<td>8317±594</td>
<td>16.63</td>
<td>299.36</td>
</tr>
<tr>
<td>S15</td>
<td>Seven Degassing station of East Qurna</td>
<td>9123±651</td>
<td>18.07</td>
<td>328.225</td>
</tr>
<tr>
<td>S16</td>
<td>Eight Degassing station of East Qurna</td>
<td>9578±684</td>
<td>18.98</td>
<td>344.78</td>
</tr>
<tr>
<td>S17</td>
<td>Zabair Degassing Station</td>
<td>32480±2320</td>
<td>64.36</td>
<td>1169.13</td>
</tr>
<tr>
<td>S18</td>
<td>Zabair musharaf Degassing Station</td>
<td>33752±2410</td>
<td>66.88</td>
<td>1214.90</td>
</tr>
<tr>
<td>S19</td>
<td>hamar musharaf Degassing station</td>
<td>40134±2866</td>
<td>79.53</td>
<td>1444.70</td>
</tr>
<tr>
<td>S20</td>
<td>alrafidia Degassing station</td>
<td>63918±4565</td>
<td>126.67</td>
<td>2301.02</td>
</tr>
</tbody>
</table>
### Table (2) Effective concentration of specific activities in a sludge model selected from the waste collection site - khadir Alma - belonging to the Thread Degassing Station of North Rumaila in the study area measured by the gamma spectrum analysis technique HPGe.

<table>
<thead>
<tr>
<th>NO</th>
<th>Radionuclide's</th>
<th>Radiation chains</th>
<th>Specific activities in Bq/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Pb-212</td>
<td>Th-232</td>
<td>56432</td>
</tr>
<tr>
<td>2</td>
<td>Bi-212</td>
<td>Th-232</td>
<td>33720</td>
</tr>
<tr>
<td>3</td>
<td>Ac-228</td>
<td>Th-232</td>
<td>52519</td>
</tr>
<tr>
<td>4</td>
<td>Pb-214</td>
<td>U-238</td>
<td>109479</td>
</tr>
<tr>
<td>5</td>
<td>Bi-214</td>
<td>U-238</td>
<td>117135</td>
</tr>
<tr>
<td>6</td>
<td>Ra-226</td>
<td>U-238</td>
<td>133851</td>
</tr>
</tbody>
</table>

| S21 | qubat safwan Degassing station | 15952±1139 | 31.61 | 574.21 |
| S22 | Zubair Warehouse-1            | 18245±1303 | 36.15 | 656.68 |
| S23 | Zubair Warehouse-2            | 20763±1483 | 41.14 | 747.29 |
| S24 | altuwa Warehouse              | 19548±1396 | 38.73 | 703.55 |
| S25 | Warehouse- ps1                | 21365±1526 | 43.90 | 797.46 |
| S26 | Warehouse in North Rumaila    | 19688±1406 | 39.01 | 708.63 |
| S27 | nahr bin Omar Degassing Station | 18216±1301 | 36.10 | 655.77 |
| S28 | majnun Degassing Station      | 91325±6523 | 180.98 | 3287.59 |
| S29 | Majnun-2 Degassing Station    | 18245±1303 | 36.15 | 656.68 |
| S30 | altuwba Degassing Station     | 30763±2197 | 60.96 | 1107.36 |
| S31 | nahr bin Omar Degassing Station | 18640±1331 | 36.94 | 662.85 |
Table (3) The effective concentrations of radium $^{226}$Ra and radium $^{228}$Ra emitted from Sludge and scale sediment models for different international studies [12]

<table>
<thead>
<tr>
<th>Country/ material</th>
<th>$^{226}$Ra kBq·kg$^{-1}$</th>
<th>$^{228}$Ra kBq·kg$^{-1}$</th>
<th>$^{232}$Th kBq·kg$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brazil -scale</td>
<td>19.1 to 323.0</td>
<td>4.21 to 235.0</td>
<td>-</td>
</tr>
<tr>
<td>Brazil -sludge</td>
<td>0.36 to 367.0</td>
<td>0.25 to 343.0</td>
<td>-</td>
</tr>
<tr>
<td>Algeria-Hard scale</td>
<td>1.0 to 950.0</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Algeria-Soft scale</td>
<td>1.0 to 300</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Tunisia-scale</td>
<td>4.3 to 658</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Algeria-slugde</td>
<td>0.069 to 0.393</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Norway-scale</td>
<td>0.3 to 32.3</td>
<td>0.3 to 33.5</td>
<td>-</td>
</tr>
<tr>
<td>Norway-slugde</td>
<td>0.1 to 4.7</td>
<td>0.1 to 4.6</td>
<td>-</td>
</tr>
<tr>
<td>Brazil-scale</td>
<td>&lt; 839</td>
<td>&lt; 377</td>
<td>-</td>
</tr>
<tr>
<td>Brazil-slugde</td>
<td>3.060</td>
<td>2.570</td>
<td>-</td>
</tr>
<tr>
<td>Various locations</td>
<td>&lt; 1,000</td>
<td>&lt; 360</td>
<td>&lt; 360</td>
</tr>
</tbody>
</table>

Fig. (4): R-222 concentration ([Bq·m$^{-3}$]) measured in Sludge samples by RAD7 in study area.
Fig. (5): The values of the specific activity of radioactive isotopes (B.kg\(^{-1}\)) in a Sludge sample and solution returning to thread Degassing Station of North Rumaila and stored at the radioactive waste collection site in Khader Al-Maa - the study area.

Fig. (6) Effective dose (Svh\(^{-1}\)) in Sludge samples in the study area.
5. **Conclusions:**

This study, for measuring the concentrations of radon gas emitted from Sludge and other radioactive isotopes from the NORM, radioactive waste collection site in the khadir alma - south of Basra city, showed Using advanced RAD electronic technology and High Purity Germanium Detectors (HPGe).

1. The largest concentration of Sludge is $98463 \pm 7031 \text{Bq.m}^{-3}$, which appeared in sample (S10) in a ±model from a sludge barrel at the study site transferred from the Thread Degassing Station of North Rumaila field, the concentration of radon gas in the sludge of the Sixth Gas Isolation Station / West Qurna field is the least concentrated of the species $8317 + 5949 \text{Bq.m}^{-3}$

2. The emergence of an increase in the concentrations of radioactive isotopes ($^{214}\text{Pb}, ^{214}\text{Bi}, ^{226}\text{Ra}$) ($^{212}\text{Pb}, ^{212}\text{Bi}, ^{228}\text{Ac}$), which belong to the uranium-238 and thorium-232 series, respectively, ibn high rates compared to their permissible concentration according to UNSCEAR.

3. It was found that the annual equivalent dose in slag samples (Sludge) in the production of slag drums studied is higher than the permissible dose level of $50 \mu\text{Sv h}^{-1}$ proposed by the EPA in the United States.

4. This concentration radon gas and the specific effectiveness of radioisotopes and doses have a negative impact on the health of the workers at the site In case of non-compliance with radiation safety procedures in the workplace.

5. The research added important information and a roadmap about radon concentrations in oil sludge at Khidir alma site for collecting and storing radioactive assemblies in Basra Governorate - southern Iraq.
References:


[10] Environmental Protection Agency of USA, A citizen’s guide to radon, what it is and what to do about it. Published with the Department of Health and Human Services, OPA-86-005 (August), 1986.


