

# Radioactivity Levels in Scales Generated from Crude Oil Production in Ghana

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## Abstract

Knowledge of accurate radio isotopic signatures is very necessary in assessing any potential radiological hazards and risks to members of the public and workers from exposure to NORM contaminated scales. In this work scales from crude oil production activities from Ghana have been assessed using alpha spectrometry after radiochemical separation, and non-destructive gamma spectrometry. Characterization and determination of activity concentrations of  $^{234}\text{U}$ ,  $^{238}\text{U}$ ,  $^{210}\text{Po}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{224}\text{Ra}$  and  $^{40}\text{K}$  have been established. The average activity concentrations of  $43.9 \pm 8.1 \text{ kBq.kg}^{-1}$ ,  $30.3 \pm 5.1 \text{ kBq.kg}^{-1}$ ,  $11.2 \pm 2.8 \text{ kBq.kg}^{-1}$  and  $11.2 \pm 2.6 \text{ kBq.kg}^{-1}$  obtained for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ , and  $^{224}\text{Ra}$  respectively in scale samples in this study exceeded the IAEA Basic Safety Standards (BSS) exemption levels giving an indication that the scale samples could present significant future radiological risk for workers, the public and environment

## Introduction

During the last decade some oil explorations and exploitation activities have started at different sites along the west coast of Ghana. Consequently, environmental concern has appeared in association with possible chemical pollution but, in addition, radiometrical characterization becomes relevant in oil extraction, which is a well-known NORM (Naturally Occurring Radioactive Material) activity.

A common problem in oil production is the accumulation of scale on the interior surfaces of production pipes. The scales are mostly composed of barium sulphate, barium carbonate, and/or calcium carbonate. NORM scale is produced when Ra dissolved in the formation water is co-precipitated with Ba, Sr, or Ca as mostly sulphates<sup>(1)</sup>. These materials form insoluble hard deposits inside the production equipment<sup>(2)</sup> and depending on the formation and age of the well, the management (removal, storage and disposal) of the scale can result in serious health and environmental risks if not handled properly, both for workers and the public.

The dominating radionuclides present in scales and other precipitates are  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , with typical concentrations ranging from 1 to 1000  $\text{kBq kg}^{-1}$ , although concentrations as high as 15,000  $\text{kBq kg}^{-1}$  have been reported in literature<sup>(3)</sup>. Values for  $^{228}\text{Ra}$  in scales are, in general, not much less than for  $^{226}\text{Ra}$ .

Radiation protection considerations arise mainly from the removal of this scale during maintenance and decommissioning operations resulting in exposure to external gamma radiation and inhalation of dust, and from the subsequent disposal of such materials as waste. Individuals working close to heavily scaled pipes and vessels may also need to be subjected to radiation protection measures.

The primary aim of the study shown in this paper has been to determine and characterize the radioactivity levels of scale generated as a result of oil and gas production in Ghana production oilfields in order to establish baseline data. The emphasis of this study is on the determination of the activity concentrations and distribution of several naturally occurring radionuclides of the U/Th decay series and  $^{40}\text{K}$  using mainly alpha-particle and high-resolution gamma-ray spectrometric techniques. This work was necessary since no studies have been carried out on radioactivity levels in scales generated as a result of oil and gas exploration and production from the oilfields in Ghana and published data are not available.

## Materials and Methods

Composite samples comprising of 5 scales were collected for this work from produced water pipes at the storage and maintenance facility onshore for the Ghana oilfields in January 2015.

The preparations for U, Th and Po determination by alpha-particle spectrometry were carried out in three main steps: pre-concentration, radiochemical separation and electrodeposition. The details of these procedures can be found in earlier presentation<sup>(4)</sup>. However, in this study the step involving calcination was excluded (due to our interest in Po for this work, the samples were wet-digested directly using aqua regia) and also a different radiochemical separation procedure using liquid – liquid solvent extraction (TBP + Xylene) according to a method described by Martinez-Aguirre<sup>(5,6)</sup> was applied to the samples. The Th fraction obtained by this separation was further purified using AGI-X8 ionic exchange resin (100-200 mesh). Source preparation for Po was done by self-deposition onto copper discs while U and Th were independently electrodeposited onto stainless steel discs by applying the well-known method of Hallstadius<sup>(7)</sup>.

The measurement of the U and Th isotopes electroplated and Po self-deposited were done using 450 mm<sup>2</sup> active surface PIPS detectors installed in an 8- chamber Alpha analyst system (Canberra). In our laboratory, each chamber is devoted to the exclusive measurement of one element in order to avoid cross contamination. The measurements were carried out at a source to detector distance of 0.5 cm. The accumulation and analysis of Alpha spectra was done using Genie 2000 software with measurement time of 200,000s. The background spectra were also used to determine the minimum detectable activity (95% confidence level) of U, Th and Po (~0.1mBq) for a measuring time of 2-3 days. The mean recovery yields of the radiochemical procedure were 69% for U, 60% in the case of Th and 70% for Po.

For gamma-ray determinations, scale aliquots were air-dried and finally oven dried in some cases to remove any additional moisture from the samples. The dried samples were each transferred into measuring cylindrical containers (55mm diameter by 12mm height) without any treatment and vacuum sealed after sealing with silicone. All samples were non-destructively analysed by high-resolution gamma spectrometry using a p-type Extended Range Germanium coaxial detector (XtRa) with relative efficiency of 37.1% and with an energy resolution of 1.8 keV for gamma-ray energy of 1332 keV of  $^{60}\text{Co}$ . The detector is housed in a 10 cm passive shielding of ancient lead with an active shielding made with an organic scintillation detector (Bicron BC-418) placed on the top of the lead shield and working in anticoincident mode with the Ge detector. This allows remarkable precision in the environmental gamma-radiation measurements due to the very low background. The counting time was 180000 seconds for each sample. The identification of individual radionuclides was performed using their characteristic gamma-ray energies and the quantitative analysis of radionuclides was performed using the Genie 2000 gamma acquisition and analysis software. Background spectra were acquired and used to correct the net peak area of gamma rays of the

measured isotopes and used to determine the minimum detectable activities of  $^{226}\text{Ra}$  (0.11 Bq),  $^{232}\text{Th}$  (0.13 Bq), and  $^{40}\text{K}$  (1.10 Bq) at the 95% confidence level.

The IAEA reference materials IAEA-RGU-1(U-ore) and IAEA-RGTh-1 (Th-ore) with mean densities ( $1.33 \pm 0.03 \text{ g/cm}^3$ ) similar to the mean densities of scale samples to be measured were prepared into same cylindrical containers as that of scale and sludge samples and were used to estimate the efficiencies for photo peaks of natural radionuclides measured and quantified in the samples.

The activity concentrations of  $^{226}\text{Ra}$  were determined using the  $\gamma$ -ray emissions and the respective  $\gamma$ -yield of  $^{214}\text{Pb}$  at 351.9 keV (35.8%) and  $^{214}\text{Bi}$  at 609.3 keV (44.8%). The  $^{228}\text{Ra}$  activity concentrations were determined through the gamma emissions of  $^{228}\text{Ac}$  at 911 keV (26.6%), and the  $^{228}\text{Th}$  activity concentrations were determined through the gamma emissions of  $^{212}\text{Pb}$  at 238.6 keV (43.3%) and  $^{208}\text{Tl}$  at 583 keV (30.1%) and 2614.7 keV (35.3%) taking into consideration a branching ratio of 33.7% from  $^{212}\text{Bi}$  towards  $^{208}\text{Tl}$ . The  $^{40}\text{K}$  activity concentration was determined directly from its emission line at 1460.8 keV (10.7%) while the  $^{210}\text{Pb}$  activity concentration was determined directly from the gamma emission line at 46.5 keV (4.3%). In calculating the activities, considerations of self-absorption correction factor for low energy photo peak  $^{210}\text{Pb}$  was incorporated. All the energies and intensities of the different radiations mentioned were taken from a well-known library <sup>(8)</sup>.

Air borne radon activity concentrations were measured directly with a Genitron Alpha Guard, Model PQ 2000/mp50. The measurements were carried out outdoor and indoor in the field at scale embedded pipes storage and maintenance facility. The temperature, atmospheric pressure and relative humidity were also recorded during the measurement. The Alpha Guard is provided with a large surface glass fibre filter, which allows only the gaseous  $^{222}\text{Rn}$  to pass through whilst the radon progeny are prevented from entering the ionisation chamber. The data was evaluated using Alpha View/ Data Expert Software, which automatically transforms radon daughter concentrations from working level (WL) to equilibrium equivalent concentration (ECC) in  $\text{Bqm}^{-3}$ .

## Results and Discussion

The results of radioactivity concentrations of radionuclides  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{224}\text{Ra}$  and  $^{40}\text{K}$  in scale samples determined by gamma spectrometry are presented in Table 1. The average value of the activity concentration of  $^{226}\text{Ra}$  was  $43.9 \pm 8.1 \text{ kBq.kg}^{-1}$  (in a range of 38.5–58.3  $\text{kBq.kg}^{-1}$ ); for  $^{210}\text{Pb}$ , the average was  $0.36 \pm 0.15 \text{ kBq.kg}^{-1}$  (in a range of 0.20–0.60  $\text{kBq.kg}^{-1}$ ); while for  $^{228}\text{Ra}$ , the average activity concentration was  $30.3 \pm 5.1 \text{ kBq.kg}^{-1}$  (in a range of 26.8 – 39.2  $\text{kBq.kg}^{-1}$ ); and for  $^{228}\text{Th}$ , the average was  $11.2 \pm 2.8 \text{ kBq.kg}^{-1}$  (in a range of 8.6–15.9  $\text{kBq.kg}^{-1}$ ). In the case of  $^{224}\text{Ra}$  the average activity concentration was  $11.2 \pm 2.6 \text{ kBq.kg}^{-1}$  (in a range of 8.8–15.4  $\text{kBq.kg}^{-1}$ ); while for  $^{40}\text{K}$ , the average is  $1.8 \pm 0.4 \text{ kBq.kg}^{-1}$  (in a range of 1.3–2.3  $\text{kBq.kg}^{-1}$ ).

Sample ID	Concentration ( $\text{kBq.kg}^{-1}$ )					
	$^{226}\text{Ra}$	$^{210}\text{Pb}$	$^{228}\text{Ra}$	$^{228}\text{Th}$	$^{40}\text{K}$	$^{224}\text{Ra}$
SC1	$38.5 \pm 0.4$	$0.4 \pm 0.01$	$26.8 \pm 0.1$	$9.8 \pm 0.8$	$1.8 \pm 0.2$	$9.6 \pm 0.9$
SC2	$41.5 \pm 0.4$	$0.3 \pm 0.01$	$28.2 \pm 0.5$	$11.0 \pm 0.6$	$1.8 \pm 0.1$	$10.9 \pm 0.8$
SC3	$40.7 \pm 0.4$	$0.2 \pm 0.01$	$27.6 \pm 0.5$	$10.9 \pm 0.5$	$1.3 \pm 0.2$	$11.2 \pm 0.7$
SC4	$40.7 \pm 0.1$	$0.3 \pm 0.01$	$29.9 \pm 0.5$	$8.6 \pm 0.6$	$2.0 \pm 0.3$	$8.8 \pm 0.6$
SC5	$58.3 \pm 0.2$	$0.6 \pm 0.1$	$39.2 \pm 0.7$	$15.9 \pm 1.2$	$2.3 \pm 0.3$	$15.4 \pm 0.9$

**Table 1.** Activity Concentration of  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  series radionuclides in Scales samples by Gamma Spectrometry.

It is clear from the data shown in Table 1, that all radionuclides except  $^{210}\text{Pb}$  compared for scale samples exceeded the IAEA Basic Safety Standards (BSS) exemption levels and hence there may be need to exert regulatory control of the NORM residues.

The results of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in scale were compared with data from other countries as shown in Table 2. The specific activities in general compare well with data from other countries around the world. The range of activity concentration of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in scale from Ghana (this study) was higher than the range of values reported in Scale from other studies carried out in other Oilfields in Norway, Congo, Egypt and Kazakhstan <sup>(9-12)</sup> and lower than those recorded in some other work undertaken in various oil fields in the United Kingdom, United States of America, Egypt, Tunisia, Algeria, Syria, Malaysia, Brazil and Australia <sup>(13-22)</sup>. The variation in activity concentration of natural radionuclide content in scales of different origins in the oil extraction industry can be attributed to factors such as the geochemical and geological characteristics of reservoir rocks, age, the type of hydrocarbons produced and operating conditions for the oilfields.

Country	$^{226}\text{Ra}$ ( $\text{kBq.kg}^{-1}$ )	$^{228}\text{Ra}$ ( $\text{kBq.kg}^{-1}$ )	Reference
Algeria	1 – 950	-	Hamlat et al., 2001
Australia	21 – 250	48 – 300	APPEA, 2002
Brazil	19.1 – 323	4.2 – 235	Godoy and Crux, 2003
Brazil	77.9 – 2110	101.5 – 1550	Gazineu and Hazin, 2008
Congo	0.097 – 0.151	-	Testa et al., 1994
Egypt	0.49 – 0.52	0.032 – 0.05	Abo-Elmagd et al., 2010
Egypt	7.5 – 143	35.46 – 368.654	Shawky et al., 2001
Ghana	38.5 – 58.3	26.8 – 39.2	Present study
Kazakhstan	0.51 – 51	0.2 – 10	Kadyrzhanov et al., 2005
Malaysia	114 – 188	130 – 207	Omar et al., 2004
Norway	0.3 – 32.3	0.3 – 33.5	Lysebo et al., 1996
Syria	147 – 1050	43 – 181	Al-Masri and Suman, 2003
Tunisia	4.3 – 658		Heaton and Lambley, 1995
UK	1 – 1000		E & P forum, 1987
USA	15 – 76		White and Rood, 2001

**Table 2.** Comparison of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in Scale and Sludge from Ghanaian oilfields with others published in literature

Table 3 presents specific activities of  $^{234}\text{U}$ ,  $^{238}\text{U}$ ,  $^{210}\text{Po}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  determined using alpha spectrometry. The activities of  $^{234}\text{U}$  were in the range  $0.9\text{--}4.5 \text{ Bq.kg}^{-1}$  with average  $1.9 \pm 1.5 \text{ Bq.kg}^{-1}$ , the activities of  $^{238}\text{U}$  in the range  $1.6\text{--}4.6 \text{ Bq.kg}^{-1}$  with average  $2.5 \pm 1.2 \text{ Bq.kg}^{-1}$ , the activities of  $^{210}\text{Po}$  in the range  $66\text{--}166 \text{ Bq.kg}^{-1}$  with average  $99 \pm 39 \text{ Bq.kg}^{-1}$ , the activities of  $^{230}\text{Th}$  in the range  $1.2\text{--}3.9 \text{ Bq.kg}^{-1}$  with average  $1.9 \pm 1.2 \text{ Bq.kg}^{-1}$  and the activities of  $^{232}\text{Th}$  in the range  $0.9\text{--}4.5 \text{ Bq.kg}^{-1}$  with average  $1.9 \pm 1.5 \text{ Bq.kg}^{-1}$  respectively.

Sample ID	Concentration ( $\text{Bq.kg}^{-1}$ )				
	$^{234}\text{U}$	$^{238}\text{U}$	$^{210}\text{Po}$	$^{230}\text{Th}$	$^{232}\text{Th}$
SC1	$2.1 \pm 0.2$	$1.6 \pm 0.1$	$97 \pm 4$	$1.3 \pm 0.2$	$0.9 \pm 0.1$
SC2	$2.9 \pm 0.3$	$2.3 \pm 0.5$	$66 \pm 3$	$3.9 \pm 0.3$	$4.5 \pm 0.3$
SC3	$3.1 \pm 0.3$	$2.4 \pm 0.3$	$83 \pm 5$	$1.3 \pm 0.2$	$1.8 \pm 0.3$
SC4	$5.5 \pm 0.4$	$4.6 \pm 0.4$	$82 \pm 2$	$1.2 \pm 0.1$	$1.1 \pm 0.1$
SC5	$2.2 \pm 0.2$	$1.8 \pm 0.2$	$166 \pm 7$	$1.7 \pm 0.2$	$1.0 \pm 0.1$

**Table 3.** Activity Concentrations of  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{U}$ ,  $^{210}\text{Po}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  in Scales determined by alpha-particle spectrometry.

The  $^{210}\text{Po}$  level was highest in all the scales samples via alpha, an indication that  $^{210}\text{Po}$  may contribute significantly to total alpha activity. This could be a significant source of exposure to workers via internal contamination by inhalation during maintenance, decommissioning operations and waste processing and disposal. However, generally activity concentrations of U and Th-isotopes as well as  $^{210}\text{Po}$  for all scale samples in this study were much lower compared to that of radium isotopes discussed earlier above.

Finally, the  $^{222}\text{Rn}$  concentrations were measured in air on the field from pipes at storage and maintenance facility and the results were  $21 - 194 \text{ Bq.m}^{-3}$  with average  $113 \pm 49 \text{ Bq.m}^{-3}$  for outdoor and  $200 - 548 \text{ Bq.m}^{-3}$  with average  $334 \pm 94 \text{ Bq.m}^{-3}$  for indoor. The airborne radon concentration measurements were made at average conditions of temperature, atmospheric pressure, and relative humidity with mean values of  $34.5 \text{ }^\circ\text{C}$ ,  $100.3 \text{ kPa}$  and  $90.4 \%$  respectively. By comparison of results, it is observed that indoor concentrations were about 3 times higher than that of outdoor measurements as a result of diffusion and dilution outdoor. This indicates that, one of the most effective means of reducing radon exposure is to provide proper and effective ventilation when the levels are high in an enclosed area. The airborne radon concentrations are on the other hand within the ICRP recommended value of  $200\text{-}600 \text{ Bq/m}^3$  in dwellings and below the value of  $1000 \text{ Bq/m}^3$  for workers <sup>(23)</sup>.

## Conclusions

Radioactive characterization of NORM contaminated scale resulting from the crude oil production activities and waste generation from oilfields of Ghana have been investigated using alpha spectrometry after radiochemical separation and non-destructive gamma spectrometry. Characterization and determination of specific activities of  $^{234}\text{U}$ ,  $^{238}\text{U}$ ,  $^{210}\text{Po}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{224}\text{Ra}$  and  $^{40}\text{K}$  for NORM residues have been established.

The specific activities of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ , and  $^{224}\text{Ra}$  for scale samples in this study exceeded the IAEA Basic Safety Standards (BSS) exemption levels and hence needs to be handled with care in the deciding its transport, removal, storage and disposal of the NORMs residues. It was also clear from this study that,  $^{210}\text{Po}$  level was highest between the radionuclides measured via alpha spectrometry, an indication that  $^{210}\text{Po}$  may contribute significantly to total alpha activity. This could be a significant source of exposure to workers via internal contamination by inhalation during maintenance, decommissioning operations and waste processing and disposal.

The airborne radon concentration measured at waste storage facilities in this study were within the ICRP recommended value of  $200\text{-}600 \text{ Bq/m}^3$  in dwellings and below the value of  $1000 \text{ Bq/m}^3$  for workers <sup>(29)</sup>.

Overall, this study suggests that removal, storage and disposal of Scale and sludge could present significant future radiological risk for workers, the public and environment if not properly managed.

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