BJRS

BRAZILIAN JOURNAL OF RADIATION SCIENCES
10-03B (2022) 01-08

Performance evaluation of NaI(Tl) and LaBr3(Ce) portable radiological identifiers in the identification of 226-Ra and 228-Ra in scales from the petroleum industry

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ABSTRACT

The scales from the oil industry are constantly being studied, due to their formation problem, which reduces the plant's production capacity, and the radioactivity in them, which originates from production wells (NORM), which can lead workers to significant exposures. The extraction process makes these scales radioactive, as the 226-Ra and 228-Ra that are mobilized precipitate with the scales. These are directly responsible for the dose increase to which workers are exposed during routine maintenance. In this study, ten scale samples were analyzed, taken from different parts of an oil production plant, using portable radiological identifiers of NaI(Tl) and LaBr3(Ce) to assess the dose rates and performance of these equipment in detection 226-Ra and 228-Ra. The identification of 226-Ra, was given by its daughter 214-Pb and 214-Bi, energies of 295.222 keV and 351.93 keV; and 609.32 keV, respectively. The identification of 228-Ra, by his daughter 212-Pb and 208-Tl, energies of 238.632 keV; and 583.187 keV and 2614.511 keV, respectively. The energy of 583.187 keV appeared only in the spectra obtained with the PRI of LaBr3(Ce). The energy of 2614.511 keV of 208-Tl was discriminated in all spectra obtained with NaI(Tl). The NaI(Tl) PRI performed better due to its better counting efficiency. The identifiers were able to identify 226-Ra and 228-Ra in inlays containing NORM. It was concluded that the PRIs allow fast analysis even in samples with little mass.

Keywords: **radioactive scales, NaI(Tl), LaBr3(Ce).**

ISSN: 2319-0612
DOI: https://doi.org/10.15392/2319-0612.2022.1872
Submitted: 2022-01-28
Accepted: 2022-09-21

1. INTRODUCTION

Studies involving scales from the oil industry are being increasingly disseminated, due to its formation can deposit in pipelines, machinery and equipments, by causing the reduction of production. Also, these scales present a certain level of radioactivity, which can lead to an increase of the dose to which workers in the oil industry are exposed during maintenance routine [1-5]. This radioactivity is due to the fact that oil wells have NORM (Naturally Occurring Radioactive Material) in their composition, from the natural radioactive series, which, during the extraction process, makes the scales to be radioactive due to the precipitation of some radionuclides that are present in produced water and barite scale, as 226-Ra and 228-Ra, produced by the radioactive decay of 238-U and 232-Th present in rocks of the oil-producing formations [6-11]. Workers who are required to work with plant or equipment which is NORM contaminated have potential for external radiation exposure from closed systems during normal operations and internal exposure if no controls are established during shutdowns and periods where systems are opened [12-13].

Measurement of total radioactivity with a handheld radiation detector permits rapid assessment of a site for NORM contamination by estimating the concentration of radium isotopes, but it requires confirmation by laboratory analysis [14-16]. Therefore, the objective of this study is to evaluate the performance of the use of portable radiological identifiers (PRI) with NaI(Tl) and LaBr3(Ce) scintillators [15], in order to previously analyze the dose rates of scales removed from different parts of the oil production plant and detect the presence of 226-Ra and 228-Ra by their descendants, showing whether it is possible to use it for the "in situ" analyzes and therefore, for a rapid decision-making on the safety of radiological protection teams in critical environments of a production plant.

2. MATERIALS AND METHODS

Ten samples, six of scales and four of sludge, named A to J, collected at different points of the process plant from a FPSO (Floating Production Storage and Offloading) platform vessel in the Campos Basin, were macerated and passed through a 0.2 mm opening, ensuring the same granulometry size. Afterwards, 25 g of the sample were weighed and placed in polystyrene containers measuring 5 cm in diameter and 2 cm in height. The integrity of the containers has been tested and guaranteed. The samples were then measured with the PRI at a distance of 5 cm from the sample/detector, as shown in Figure 1. Since the purpose of using these equipments is the field analysis for rapid identification of radionuclides and dose rate verification [17], it is not necessary to ensure the secular balance.

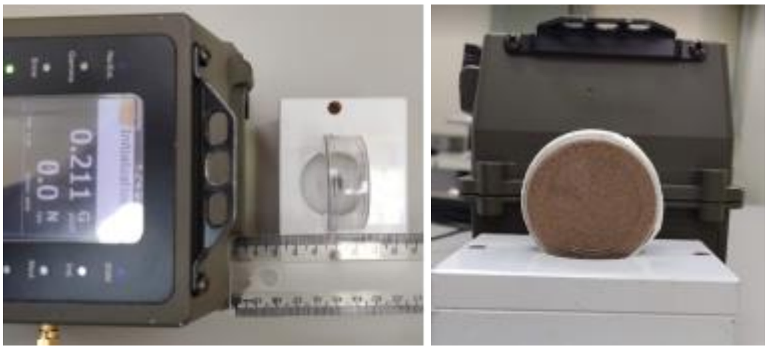

Figure 1: *Correlation geometry*
Source: Author

The PRI with scintillators of NaI(Tl) and LaBr3(Ce) used in this study have 3.0" x 1.5" crystal with 7.5% resolution and 1.5" x 1.5" crystal with 2.9% resolution, respectively. Resolutions are referenced to 137-Cs. They operate with a measurement range from 25 keV to 3000 keV and a dose rate of 0.01 µSv/h to 10 mSv/h, programmed for acquisition in 180 seconds, presenting energy distribution spectra in 1024 channels, and maximum dose rate measured during this acquisition time [17]. PRI are equipment calibrated by the manufacturer. Therefore, whenever a new set of measurements is started, its calibration is verified using certified sources [18].

3. RESULTS AND DISCUSSION

The dose rates obtained with LaBr3(Ce) showed themselves slightly than those obtained with the NaI(Tl), as shown in Table I, but without changing its order of magnitude, presenting a maximum difference elevation of 0.07 µSv/h between the results.

Table 1: Dose rates obtained with PRI.

	Portable Radiologic Identifiers (μSv/h)	
Sample	**NaI(Tl)**	**LaBr3(Ce)**
A	0.34	0.39
*B	0.25	0.29
C	0.32	0.39
*D	0.30	0.31
E	0.21	0.26
*F	0.31	0.32
G	0.30	0.34
H	0.31	0.36
I	0.23	0.24
*J	0.25	0.27
BG	0.20	0.23

*Sludge sample

Table 2 shows the energies that each PRI was able to identify, indicating the isotopes detected in each sample.

Table 2: Energies identified by PRI for each sample

Isotope	**Energy (keV)**	**PRI NaI(Tl)**	**PRI LaBr3(Ce)**
212-Pb	238.632	A,C,D,F,G,H,I	A,C,F,G,H,I,J
214-Pb	295.222	C,D,F,G,H	A,B,C,D,F,G,H

214-Pb	351.932	A,B,C,D,F,G,H	A,B,C,D,F,G,H,J
208-Tl	583.187	--	A,B,E
214-Bi	609.320	A,B,C,D,E,F,G,H,I,J	C,D,F,G,H,I,J
208-Tl	2614.511	A,B,C,D,E,F,G,H,I,J	C,H

The identification of 226-Ra, was given by its daughter nuclides 214-Pb and 214-Bi, by the energies of 295.22 keV and 351.93 keV; and 609.32 keV, respectively. And the identification of 228-Ra, by his daughter nuclides 212-Pb and 208-Tl, by the energies of 238.632 keV; and 583.187 keV and 2614.511 keV, respectively. The energy of 583.187 keV appears only in the spectra obtained with the PRI of LaBr3(Ce), but it can be observed in Table II that when this energy appears, the energy of 609.32 keV does not appear. That is, as they are close energies, with only 26 keV of difference, it can be assumed that due to the energy resolution of the equipment, it cannot discriminate the two energies. The same occurs with the NaI(Tl) that presents a worse resolution, compared to the LaBr3(Ce), which opens the base of the gaussian preventing the discrimination of these energies. The 2614.511 keV energy of the 208-Tl was discriminated in all spectra obtained with NaI(Tl) due to its better counting efficiency, even with low counting statistics. As it is a high energy, it is influenced by smaller energies, widening its gaussian shape, which makes difficult a correct analysis of the peak area. However, its identification confirms the presence of 232-Th in the sample [15], as expected.

4. CONCLUSION

The two identifiers are able to identify 226-Ra and 228-Ra in NORM containing scales, thus, both can be used for in situ analysis. However, the PRI of NaI(Tl) presented better performance because it has a better counting efficiency, due to its crystal has a larger area than the $LaBr_3(Ce)$ one, offering a faster response, almost immediate, even with low counting statistics, mainly for well defined energies. Showing that larger active volumes register more signals than smaller active volumes, under the same conditions and measurement geometry.

The PRI of $LaBr_3(Ce)$ despite presenting better resolution, during the measurements it took approximately 10 seconds to give a response, that is, the crystal geometry and the low counting statistic affect the equipment response time. As they are equipments with different types and sizes of crystals, the dose rates were slightly different, but without changing their order of magnitude. It is concluded that the PRI allows the fast identification of NORM even in samples with low mass.

ACKNOWLEDGMENT

The authors would like to thank to the Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) and Fundação de Amparo à Pesquisa do Estado do Rio de Janeiro (FAPERJ) for the financial support.

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