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Evaluation of radiological impacts of tenorm in the Tunisian petroleum industry

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ABSTRACT

The health impacts associated with uncontrolled release of TENORM in products and wastes released in the petroleum industry are of great concern. In this study, evaluation of TENORM in the Tunisian petroleum products and wastes is presented. Fourteen products samples, twelve waste samples and three samples from the surrounding environment were collected from the Tunisian Refinery STIR site and from two onshore production oilfields. The activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K for all samples were determined using gamma-ray spectrometry with High Purity Germanium (HPGe) detector. The activity concentrations of ^{224}Ra were calculated only for scale samples. The radium equivalent activity, external and internal hazard indices, absorbed doses rates in air and annual effective dose were also estimated. It was noticed that maximum value of Ra_{eq} activity was found to be 398 Bq/kg in scale (w8) collected from an onshore production oilfield which exceeds the maximum Ra_{eq} value of 370 Bq/kg recommended for safe use. All hazard indices indicated that scale samples (w6, w7, w8 and w11) could be a significant waste problem especially sample (w8). In this study, the radium isotopic data were used to provide an estimate of scale samples ages by the use of the ^{224}Ra/^{228}Ra activity ratio dating method. Ages of collected scales were found to be in the range 0.91–2.4 years. In this work, radioactivity (NORM contamination) in samples collected from the refinery STIR are showed to be insignificant if compared to those from onshore oilfield production sites.

1. Introduction

The awareness about radiation doses due to natural radiation has been cause of increased concern in recent years. In this context, particular attention has been focused on the mining and processing industries, the latter being generally evocated for the presence of Naturally Occurring Radioactive Materials (NORM) in products or residues (Gazineu and Hazin, 2008).

The presence of higher than background radioactivity in crude petroleum was reported for the first time, more than a century ago, by Himstedt (1904) and Burton (1904). In the 1920–1930's, the presence of NORM was also reported in numerous Russian research papers, however the first official survey, from a radiation protection point of view, was not done until the early 1970's (AEC, 1972).

Later reports described the occurrence of ^{226}Ra in reservoir water from oil and gas fields (Kolb and Wojcik, 1985; Born, 1936) and in the 1970's and 1980's several observations impelled renewed interest (Gesell, 1975; Summerlin and Prichard, 1985; Kolb and Wojcik, 1985; API, 1992; Al-Masri et al., 1997; Raeburn et al., 1988; Otto, 1989; UKOOA, 1985; Waldram, 1988).

The presence of elevated NORM concentrations (i.e., above background) in some oil and gas waste has been recognized since the early 1930s (Otto, 1989); however, NORM concentrations have been largely unregulated. Since the mid-1980s, both federal and state regulatory agencies have become increasingly concerned about the presence of NORM.

In the petroleum industry, NORM such as the ones from the ^{238}U and ^{232}Th series, as well as ^{40}K are often enhanced as a result of industrial operations, these materials are formally referred to as Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM).

Whilst the activity levels of these NORM are not always enhanced, simple chemical or physical changes can sometimes take place, resulting in radionuclides being more readily available for transfer by various pathways (Heaton and Lambley, 1995; UNSCEAR, 2001).

Naturally Occurring Radioactive Materials (NORM) resulting from the ^{232}Th and ^{238}U-series can be concentrated and accumulated in tubing and surface equipment in the form of scale and sludge as a consequence of physical and chemical processes

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associated with the oil and gas industry (IAEA, 2004; Jonkers et al., 1997; API, 1992).

In addition, produced water contains enhanced naturally occurring radioactive materials (NORM) resulting from the ^{232}Th and ^{238}U series (API, 1992; Baird et al., 1996; IAEA, 2004; Jonkers et al., 1997; Shawky et al., 2001; NRPA, 2004; Swann et al., 2004); this water is currently considered to be the largest volume of radioactive waste generated by the oil industry (IAEA, 2004).

Although the initial production of oil and gas from a reservoir is typically "dry", formation waters may be increasingly produced along with the oil and gas as the natural pressure within the petroleum-bearing formation decreases. As produced waters are brought to the surface, decreases in temperature and pressure allow solutes contained within the waters to precipitate (Smith, 1987). This can result in the formation of hard, extremely insoluble barite scale deposits on the interior surfaces of piping, on casing materials, and on other production equipment. In the case of piping, these deposits may eventually restrict flow sufficiently to require replacement of the scaled pipe segments (Raabe, 1996; Bernhardt et al., 1996).

Such scale, accumulated in production pipes may be radioactive. This radioactivity is due to radium co-precipitating with barium and strontium sulfates in the scale formation. Since uranium and radium have different chemical properties, they are affected to a different extent by chemical leaching and do not necessarily concentrate in the same manner (Durrani and Ilic, 1997).

The radioactivity of scale formed in oil and gas production processes is an important issue, particularly from the point of view of radiation protection and has recently received considerable attention (Testa et al., 1994).

The aim of this work is to explore the incidence of radioactivity in the Tunisian petroleum industry (refining and oil production) since, to the best of our knowledge, no studies have been done on this subject.

The present paper consists of the determination of activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K content in the petroleum products and wastes collected both from the Tunisian refinery STIR and from two onshore production oilfields sites (site N°1: situated in the south of Tunisia and site N°2: located in the north of Kairouan concession, approximately 190 km south of Tunis) presented on Fig. 1. The radium equivalent activity, radiation hazard indices,

Fig. 1. Geographic situation of oil field production site N°1 (Oued Zar field) and oil field production site N°2 (Sidi Kilani field).