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NORM and the Brazilian Non-Nuclear Industries

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ABSTRACT

It is well known that there are areas of high natural radioactivity in Brazil. Among the best known are Guaraparí, Poços de Caldas and Araxá. However, there are also a number of lesser known areas with orebodies bearing commercially important minerals associated with thorium and uranium. Some of those areas became sites of extractive industries that produce a variety of minerals plus significant amounts of naturally occurring radioactive materials (NORM) wastes, as byproducts. Rare earths rich monazite sands can be found in Buena, Cumuruxatiba, Mato Preto and Sapucai, in addition to Guaraparí. In Gandarela and Jacobina there are gold bearing minerals associated with uranium. Extraction of copper are taking place in Salobo area of the Carajás mining complex, where typical uranium concentrations are of the order of 1000 ppm U_3O_8. It is estimated that the chemical attack of the phosphate ores of Araxá, Tapira and Catalão will leave behind NORM wastes with concentrations of at least 3kBq ^{226}Ra/kg. The pyrochlore operations in Araxá produce wastes with ^{226}Ra concentrations of the order of 100 kBq/kg; being the ^{228}Ra concentrations one order of magnitude higher. The mining area of Pitinga produces tin, tantalum, and niobium with large amounts of NORM wastes as byproducts. The paper will briefly describe the history of NORM wastes produced in the early days of the studies on natural radioactivity, and will also present an overview of the current NORM wastes situation in selected Brazilian non-nuclear industries. Environmental and regulatory implications will be examined vis-avis the Basic Safety Standards for Protection against Ionizing Radiations and for the Safety of Radiation Sources, approved at the 847th Meeting of the Board of Governors of the International Atomic Energy Agency on 12 September 1994, and adopted and/or approved by five other well regarded international organizations.

INTRODUCTION

Today it is known that the natural radioactivity extant in the Earth has its origin associated with the nucleosynthesis in stars. The phenomenon of nucleosynthesis "*can occur within the framework of any cosmology, and in particular in either the explosive-evolutionary or the steady-state cosmologies*" (Fowler, 1967). Notwithstanding, the sequence of events that started with the Röentgen discovery of the x-rays, in Wurzburg (Glasser, 1934; Dibner, 1968; Cassette, 1996), and lead to the discovery of the phenomenon of radioactivity, occurred only about 100 years ago (Becquerel, 1896a,b; Curie, 1898a, b,c).

Uranium, which played an important role in the episodes of the discovery of radioactivity, was known to exist since 1784, ans started being used in arts and crafts as UCl_4 in 1841 (Paschoa, 1997a). However, only after World War II became important again for strategic, political and economical reasons.

Marie and Pierre Curie ordered large amounts of tailings from the glass and ceramic industry from Bohemia, then part of the Austrian -Hungarian Empire, to separate radium for their experiments (Tesiska, 1989; Thomas, 1990a, b; Keller, 1993; Thomas, 1993). The first amounts of naturally occurring radioactive materials (NORM) wastes were produced by the Curies experiments. It was estimated that a total of about eight tons of raddium-bearing tailings were sent to the Curies (Landa,

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1993), with ^{226}Ra concentrations roghly between 1 and 10 Mbq/kg (Thomas, 1993). It could be instructive to examine the fate of the NORM wastes left behind by the Curies.

Thorium was discovered by the Swedish chemist Baron Jöns Jakob Berzelius, in Lövön, Norway (Cuthbert, 1958), and it is known to be present in monazite since 1839 (Paschoa, 1994). After the simultaneous discovery of the radioactivity of thorium in Paris, France (Curie, 1898a), and in Erlangen, Germany (Schimdt, 1898), the transmutation of thorium in thorium X (^{224}Ra) and thorium emanation (^{220}Rn) (Rutherford, 1900; 1902a), and subsequently the chemical separation of thorium was achieved (Rutherford, 1902b). The latter allowed physicians to start using thorium progeny componds for therapeutical applications, as a substitute for the more expensive ^{226}Ra (Badash, 1979). It is quite likely that thorium progeny bearing wastes were produced with those earlier therapeutical applications. However, thorium was used industrially even before the discovery of radioactivity. In 1885, the Austrian Baron Carl Auer von Welsbach patented a process to make a fabric bag impregnated with a mixture of thorium, lanthanum and cerium, that enhanced the brightness of a glass flame (Cuthbert, 1958). At that time an Englishman was transporting clandestinely tens of tons of monazite from Brazil to Europe (Paschoa, 1994, motivated also by the early belief that monazite was an aggregate to some diamondiferous minerals (Leonardos, 1955). Chemical processing of monazite was not introduced until the forties (Leonardos, 1951). NORM wastes bearing mesothorium (^{228}Ra) started being produced in Brazil ever since.

This paper will examine to the extent possible the current and potential situation of the NORM wastes produced by the mineral extraction and other industries that deal, sometimes without knowing, with materials bearing naturally occurring radioactive components.

NORM AND MONAZITE

The traditional methods to treat monazite to produce thorium, uranium and rare earths are all derived from a few (Kremers, 1949; Rohden, 1951; Leonardos, 1951; Bearse, 1954). By and large, the methods are composed of two phases: (i) physical separation that consists essentially in extracting, washing and drying the monazite bearing sands, which are then concentrated and separated electrostatically and electromagnetically to ; and (ii) the mineral concentrates obtained in the first phase — ilmenite, rutile, monazite, and zirconite — are further treated physically and chemically to obtain a series of purified materials for industrial applications. A more detailed description of the treatment of the Brazilian monazite can be found elsewhere (Nouailhetas, 1993; Paschoa, 1993; 1994).

Typical ranges of activity concentrations found in soils contaminated NORM wastes from the monazite cycle in Brazil are 0.3 - 33 kBq ^{228}Ra/kg and 0.06 - 6.7 kBq ^{226}Ra/kg (Nouailhetas, 1993). The main problem with the NORM wastes is rather the large volume and mass distributed in trenches and drums than the activity concentrations in locally contaminated soils. The latter can be decontaminated, though at significant cost. However, a solution must be find for the hundred of thousands of metric tons of NORM wastes that extant in non-appropiate sites. The ^{228}Ra and ^{226}Ra activity concentrations in the monazite cycle in Brazil increase by factors of about 20 thousands and 10 thousands, respectively. Even if a change in industrial procedures occurs, the need to find a solution for the NORM wastes from the monazite cycle in Brazil remains.

NORM AND PYROCHLORE

The Brazilian pyrochlore — $RNb_2O_6.R(Ti,Th)O_2$ — reserves in Araxá amount to about $4.6x10^8$ metric tons with 2.5% Nb_2O_5 as an average. The installed capacity allows a production rate of about $2.5x10^4$ metric tons/year. This means that taking into account the present demand for niobium, the Araxá reserves alone could last for a few centuries. One can estimate, based on the current industrial processes, and on the activities concentrations of the neutral water layer that cover the solid wastes, that in those wastes the ^{226}Ra and ^{228}Ra concentrations range from 0.2 to 100, and from 0.5 up to 300 kBq/kg, respectively Paschoa, 1981; Paschoa, 1997b).

NORM AND APATITE

The apatite from Araxá and Tapira, in Minas Gerais, and from Catalão, in Goiás, are ground and separated magnetically, before being chemically attacked to form P_2O_5 to be used as phosphate fertilizer. A more detailed account of the industrial process used to produce phosphate concentrates from Brazilian apatite is presented elsewhere (Paschoa, 1981). However, it is estimated that the NORM wastes resulting from this industrial process are larger than 3 kBq/kg, for both, ^{228}Ra and ^{226}Ra (Paschoa, 1981; 1997b).

NORM AND TIN

Data available from the Pitinga mine in the northernmost part of Brazil do not allow to estimate the concentrations of ^{228}Ra and ^{226}Ra in the NORM wastes left behind by the annual production of 13 kt of tin concentrate, 17 kt Ta/Nb, 68 kt of zirconite, and 500 t U_3O_8 . However, it is estimated that more than 10 kt of wastes containing NORM in a variety of degrees will be produced annually in that mine.

NORM AND OTHER MINERALS

Among the ore bodies containing uranium and/or thorium associated with other commercially feasible for the extractive industries, one can mention: gold in Gandarela and Jacobina, copper in the Salobo area of the Carajás mineral province, tin and tantalum in Ariquemes, and lead and silver in Boquira (Mafra Guidicini, 1991). All those extrative operations will produce NORM wastes in greater or lesser degree, depending on the industrial processes. Some extractive industries are not aware of the possibility of producing significant amounts of NORM wastes. As a consequence, no radioprotection is maintained in the operations. Thus, their operators are at greater risk tahn necessary.

NORM AND THE PETROLEUM INDUSTRY

The petroleum industry started being aware of the implications that oil and gas field wastes contaminated with NORM may have in their day-to-day operations. However, the main attention is given to the possibility of losses in the oil and gas production. It has been estimated that in the United States alone production losses of up to 20% in oil and 8% in gas could occur by the year 2000, if very stringent regulations would be adopted for NORM (Smith, 1995). The origins of NORM in oil and gas have received relatively meager attention (Smith, 1992), but at least one author has attempted to relate NORM with petroleum origin (Paschoa, 1997c).

The Brazilian petroleum industry produces more than 60% of the oil from offshore plataforms. The amount and activity concentrations of NORM produced in Brazil by the industry of petroleum is not available. However, there is no reason to expect that the typical levels will be much different from elsewhere in the world. Thus, it is reasonable to hypothsize that levels around 10 kBq ^{228}Ra/kg and 30 kBq ^{226}Ra/kg can be found in scales and sludge. The orders of magnitude of the actual activity concentrations cn easily be one or two orders of magnitude higher or less, but it is quite unlikely that there is not any NORM wastes from the Brazilian petroleum industry.

NORM AND THE BSS

NORM wastes were not specifically contemplated in the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, better known as BSS (IAEA, 1994). However, considering IAEA established the principles for the exemption of radiation sources and practices from regulatory control (IAEA, 1988), exempt activity concentrations (EACs) of 10 kBq/kg for both, ^{228}Ra and ^{226}Ra, can be adopted at least as a starting point for regulating NORM (Paschoa, 1996).

Table 1 presents for comparison purposes, the EACs for ^{228}Ra and ^{226}Ra, and activity concentrations found or estimated in selected NORM wastes in Brazil.

Table 1. Exempt activity concentrations (EACs) for ^{228}Ra and ^{226}Ra, and activity concentrations found or estimated in selected NORM wastes in Brazil.

Radionuclide	Activity concentration (kBq/kg)				
	EAC	Monazite	Pyrochlore	Apatite	Oil
Ra-226	10	0.06 - 6.7	0.2 - 100	3	30
Ra-228	10	0.3 - 33	0.5 - 300	3	30

Taking into account the comparison shown in Table 1, NORM wastes from apatite would be the only one type exempted from regulations. However, one must bear in mind the following: (i) Table 1 has no validity whatsoever, as far as regulating NORM is concerned; and (ii) a solution must be found for those NORM wastes which exceed the EACs, whatever they will be.

CONCLUDING REMARKS

1. The knowledge of the fate NORM wastes left behind by the Curies and other pioneers in experimental studies on natural radioactivity might be instructive for today's world decision makers.

2. Several decades of extraction and treatment of thorium and uranium bearing minerals in Brazil produced in Brazil in certain cases, like monazite and pyroclore, large amounts of NORM wastes with activity concentrations for ^{228}Ra and ^{226}Ra that reach tens and hundreds of kBq/kg.

3. Industries that deal with uranium and/or thorium in association with valuable metals — like gold, silver or copper — will have to pay attention to their industrial processes and procedures to minimize the production of NORM wastes, to improve the radioprotection of their workers, and to preserve the environment from radioactive contamination.

4. The petroleum industry will have to be regulate somehow, as far as the production and management of NORM is concerned.

5. The EACs for ^{228}Ra and ^{226}Ra may be a satisfactory starting point to regulate NORM wastes.

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