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ARTICLE

Radionuclide content of, and radiological hazards associated with, samples from the different streams of metal recycling facilities

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Abstract – Radioactive materials in scrap metals are a radiation protection problem and can become a serious concern when melted in metal recycling facilities. In this study, gamma spectroscopic measurements were carried out on waste from a major metal recycling facility. The activity concentrations (Bq.kg^{-1}) of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in the samples ranged from 3.60 ± 0.71–38.05 ± 2.29, 7.07 ± 2.16–80.42 ± 10.67, 6.89 ± 1.98–179.22 ± 6.00 and 1.69 ± 0.22–2477.76 ± 9.58, respectively. The dose rate and annual effective dose ranged from 2.34–134.75 nGy.h^{-1} and 3.28–188.64 μSv, respectively. The calculated radium equivalent index values were less than 370 Bq. The implications of these results are discussed.

Keywords: Scrap metal / Radioactive contamination / Activity concentration / Effective dose

1 Introduction

Waste generated from recycling radioactively contaminated scrap metals is a radiation protection concern for personnel and the public (IAEA, 2012) due to the health implications of exposure to radiation. The elevated radiation levels in the environment reported in many studies carried out on soil and water samples (UNSCEAR, 2008) have been attributed to mainly human activities including metal recycling (Abo-Elmagd *et al.*, 2010; Landsberger *et al.*, 2013).

Radioactivity measurements from these human activities should therefore receive the attention of research communities and regulatory authorities. However, while there have been some reported studies on radioactivity measurement in mining and oil industries, those for scrap metal facilities are very scanty (Kreh and Dewji, 2008; Trotti *et al.*, 2008; Cantaluppi *et al.*, 2012).

In the present work, which is to show that global attention still needs to be focused on radioactivity in recycling facilities, gamma spectroscopic measurements were carried out on samples collected from the different streams of a major scrap metal recycling facility.

2 Method

The facility used for the study is an electric arc furnace metal recycling facility, with reports of a high background radiation level, in the Niger Delta Region of Nigeria. Representative samples were collected at the input stream (mixture of soil and scrap metal particles), process stream (near the conveyor belt) and waste streams (slag and furnace dust) of the facility. The samples were dried, ground and then sieved with a 5-mm mesh sieve. About 650 g of each sample were packed into a Marinelli beaker, tightly closed and sealed, and stored for more than one month to ensure secular equilibrium. Gamma spectrometric measurements were carried out using an ORTEC high-purity germanium detector. The energy and efficiency calibration of the detector was carried out using a 650-g mixed CANBERRA soil standard packed in a Marinelli beaker.

The activity concentration was determined using

$$A = \frac{N}{\varepsilon \times y \times M \times T} \quad (1)$$

where A, N, ε and y are the activity, net count, efficiency and emission probability of the radionuclide, respectively. M is the mass of the sample and T is the counting time (36000 s). Selected gamma lines of ^{228}Ac and ^{208}Tl were used to determine ^{232}Th, while ^{214}Bi and ^{214}Pb were used for ^{238}U. Energy lines of 1460.8 keV and 661.7 keV were used for ^{40}K and ^{137}Cs, respectively. The errors associated with the activity concentrations were estimated using

$$\Delta A = A \times \sqrt{\left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta \varepsilon}{\varepsilon}\right)^2 + \left(\frac{\Delta y}{y}\right)^2}. \quad (2)$$

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Table 1. Activity concentration (Bq.kg^{-1}) of the radionuclides in the waste samples.

Stream	Sample Type	^{238}U	^{232}Th	^{40}K	^{137}Cs
Waste	Furnace dust	8.69 ± 2.81	7.07 ± 2.16	62.91 ± 3.58	1400.58 ± 5.92
		13.79 ± 2.56	15.61 ± 2.64	64.21 ± 3.73	436.97 ± 2.56
		10.28 ± 2.22	10.95 ± 2.83	31.54 ± 2.82	939.39 ± 4.29
Waste	Slag	8.22 ± 0.98	9.89 ± 1.57	7.62 ± 1.59	19.18 ± 0.48
		8.91 ± 1.28	10.50 ± 2.10	65.56 ± 3.82	89.63 ± 0.95
Input	Soil/metal particles	10.40 ± 1.24	13.75 ± 1.93	24.77 ± 2.49	55.37 ± 0.75
		19.31 ± 2.18	22.41 ± 2.51	12.66 ± 3.81	18.05 ± 0.58
Process	Soil/metal particles	3.60 ± 0.71	8.33 ± 1.10	6.89 ± 1.98	1.69 ± 0.22
		38.05 ± 2.29	80.42 ± 10.67	89.05 ± 5.30	54.26 ± 0.98
Mould	Furnace Dust	8.04 ± 3.72	10.52 ± 1.84	179.22 ± 6.00	2477.76 ± 9.58
	AVERAGE	12.93 ± 2.00	19.00 ± 2.94	54.44 ± 3.51	NC
	MINIMUM	3.60 ± 0.71	7.07 ± 2.16	6.89 ± 1.98	1.69 ± 0.22
	MAXIMUM	38.05 ± 2.29	80.42 ± 10.67	179.22 ± 6.00	2477.76 ± 9.58

NC = Not calculated because of the very large range in values.

The annual effective dose (E) to personnel from external exposure was calculated using

$$E = DR \times OF \times CF. \quad (3)$$

OF is the amount of time spent in the contaminated area and assumed to be 2000 hr.yr^{-1}. CF is the absorbed dose in air to effective dose conversion factor and is taken to be 0.7 Sv.Gy^{-1} (UNSCEAR, 2008). DR, which is the absorbed dose rate, was calculated using

$$DR = \alpha A_K + \beta A_U + \gamma A_{Th} + \delta A_{Cs}. \quad (4)$$

Depending on the surface area of the waste heaps facing locations where personnel work, the values of α, β, γ and δ were taken from Markkanen (1995). The contribution of U and Th to the committed effective dose (E_C) of the workers was estimated using (UNSCEAR, 2008)

$$E_C = (A_U e_U + A_{Th} e_{Th})\, D_L B_r. \quad (5)$$

e_U and e_{Th} are the uranium and thorium effective dose coefficients, respectively. D_L and B_r are the dust loading factor and breathing rate, respectively. A dust loading of 1 mg.m^{-3}(IAEA, 2006) is assumed. e_U, e_{Th} and B_r were taken to be 2.9 (55) μSv.Bq^{-1}, 25 (68) μSv.Bq^{-1} and 7300 m^3.yr^{-1} (UNSCEAR, 2008). The radium equivalent index (Ra_{eq}) was calculated using (Zaidi *et al.*, 1991):

$$Ra_{eq} = A_U + 1.43 A_{Th} + 0.077 A_K. \quad (6)$$

3 Results and discussion

3.1 Activity concentrations

The activity concentrations are presented in Table 1 for ^{238}U, ^{232}Th and ^{40}K, with averages of 12.93 ± 2.00, 19.00 ± 2.94 and 54.44 ± 3.51 Bq.kg^{-1}, respectively. The concentrations are within previously reported values for soils and their corresponding average values are lower than the world average concentration of 37 ± 4 for ^{238}U, 33 ± 3 for ^{232}Th and 400 ± 24 for ^{40}K (UNSCEAR, 2008). All the values are also less than the clearance levels of 1000, 1000 and 10000 Bq.kg^{-1} (IAEA, 2012) recommended for ^{238}U, ^{232}Th and ^{40}K, respectively. This suggests that the activities of the metal recycling facility need not be regulated. The activity of ^{137}Cs ranged from 1.69 ± 0.22–2477.76 ± 9.58 Bq.kg^{-1}. Some of the values were greater than 100 Bq.kg^{-1}, above which scrap metal recycling is considered unsafe (IAEA, 2012). In agreement with previous reports (IAEA, 2012), activities of ^{137}Cs in dust samples are relatively higher than in other samples. Table 2 shows that the activity of the NORM in the dust and slag samples was within the range reported in previous studies (Kontol *et al.*, 2008; Trotti *et al.*, 2008).

Table 2. Comparison of NORM activities (Bq.kg^{-1}) with literature values.

Sample	Radionuclides			Reference
	^{238}U	^{232}Th	^{40}K	
Dust	3.0–27.0	0.3–11.0	<3.0–6219.0	Trotti *et al.* (2008)
	8.0–13.8	7.1–15.6	31.5–179.2	This study
Slag	950	420	–	Kontol *et al.* (2008)
	8.2–8.9	9.9–10.5	7.6–65.6	This study

3.2 External dose rates and annual effective dose

The dose rates due to NORM presented in Table 3 have a mean value of 10.81 ± 7.17 nGy.hr^{-1}, which is below the world average values (50–59 nGy.hr^{-1}) (UNSCEAR, 2008). The annual effective doses ranged from 0.003–0.032 mSv. This shows that the effective doses are less than the world average value (0.066 mSv) (UNSCEAR, 2008). Dose rates due to ^{137}Cs

Table 3. Radium equivalent, dose rates, effective doses and the committed effective dose to the facility's personnel.

Stream	Sample type	Ra_{eq} Bq.kg^{-1}	DR(NORM) nGy.hr^{-1}	H(NORM) μSv.yr^{-1}	DR(Cs) nGy.hr^{-1}	H(Cs) μSv.yr^{-1}	DR nGy.hr^{-1}	H μSv.yr^{-1}	C. Eff Dose μSv.yr^{-1}
Waste	Furnace dust	23.60–40.95 (30.95)	3.62-8.72 (6.14)	5.06-12.21 (8.59)	22.72–103.33 (66.3)	31.81–144.67 (92.81)	28.79–112.05 (72.43)	40.32–156.87 (101.4)	1.47–3.14 (2.27)
Waste	Slag	22.86–28.89 (25.878)	9.81-12.92 (11.37)	13.74–18.09 (15.92)	3.07–14.34 (8.71)	4.30–20.08 (12.19)	12.88–27.26 (20.07)	18.04–38.17 (28.1)	1.98–2.10 (2.04)
Input	Soil/metal particles	31.87–52.17 (42.02)	13.77–22.38 (18.07)	19.27–31.33 (25.3)	2.89–8.86 (5.87)	4.04–12.40 (8.22)	22.62–25.27 (23.94)	31.67–35.37 (33.52)	2.73–4.50 (3.61)
Process	Soil/metal particles	15.99–159.35 (87.67)	2.26–22.62 (12.44)	3.16–31.66 (17.41)	0.09–2.82 (1.45)	0.12–3.95 (2.04)	2.34–25.44 (13.89)	3.28–35.61 (19.45)	1.60–15.48 (8.54)
Mould	Furnace dust	37.62	5.9	8.26	128.84	180.38	134.75	188.64	2.19
AVERAGE		44.16	10.81	15.13	35.98	50.37	46.79	65.50	3.74
MINIMUM		15.99	2.26	3.16	0.09	0.12	2.34	3.28	1.47
MAXIMUM		159.35	22.62	31.66	128.84	180.38	134.75	188.64	15.48

( ) mean value.

ranged from 0.09–128.84 nGy.hr^{-1}, and the annual effective doses ranged from 0.0001–0.18 mSv. The total (NORM+Cs) annual effective dose ranged from 0.003–0.189 mSv. The average value (0.0655 mSv) is about the same as the world average value, but below values (0.08–0.30 mSv) reported by Kreh and Dewji (2008) for workers of a recycling facility.

3.3 Committed Effective Dose and Radium Equivalent Index

Committed effective doses from annual intake of U and Th, presented in Table 3, ranged from 1.47–15.48 μSv.yr^{-1}. These values are less than the committed effective doses (2.85–5.60 mSv.yr^{-1}) reported in the phosphate mining industry, but greater than the maximum effective doses (<0.4 μSv.yr^{-1}) from a steel production facility (UNSCEAR, 2008). Ra_{eq} presented in Table 3, ranged from 15.99–159.35 Bq.kg^{-1}. The Ra_{eq} values were below the recommended value of 37 Bq.kg^{-1} (Zaidi *et al.*, 1991). Therefore, the inclusion of the waste in building materials will pose no unacceptable radiological risk. It should, however, be used with caution because of the presence of ^{137}Cs.

4 Conclusion

NORMs and ^{137}Cs were detected, with ^{137}Cs at a level above the metal recycling clearance level. The calculated total dose rate was greater than the world average. Except for the presence of ^{137}Cs, it is radiologically safe to include the waste in building material. The levels of ^{137}Cs suggest the need for increased regulatory supervision of metal recycling activities, especially in developing countries.

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