Research Paper https://doi.org/10.7733/jnfcwt.2023.043

Development of a Methodology for Estimating Radioactivity Concentration of NORM Scale in Scrap Pipes Based on MCNP Simulation

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(Received November 16, 2023 / Revised November 30, 2023 / Approved December 18, 2023)

Concerning the apprehensions about naturally occurring radioactive materials (NORM) residues, the International Atomic Energy Agency (IAEA) and its member nations have acknowledged the imperative to ensure the radiation safety of NORM industries. Residues with elevated radioactivity concentrations are predominantly produced during NORM processing, in the form of scale and sludge, referred to as technically enhanced NORM (TENORM). Substantial quantities of TENORM residues have been released externally due to the dismantling of NORM processing factories. These residues become concentrated and fixed in scale inside scrap pipes. To assess the radioactivity of scales in pipes of various shapes, a Monte Carlo simulation was employed to determine dose rates corresponding to the action level in TENORM regulations for different pipe diameters and thicknesses. Onsite gamma spectrometry was conducted on a scrap iron pipe from the titanium dioxide manufacturing factory. The measured dose rate on the pipe enabled the estimation of NORM concentration in the pipe scale onsite. The derived action level in dose rate can be applied in the NORM regulation procedure for on-site judgments.

Keywords: NORM, TENORM, NORM residue, Residual radioactivity, In situ measurement

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Fig. 1. The scales in the discharged scrap iron pipes during maintenance period of the TiO_2 processing factory.

1. Introduction

Some raw materials contain naturally occurring radioactive materials (NORM) and they are processed to manufacture industrial material. The ^{238}U and ^{232}Th decay series and ^{40}K are the main interest in the scope of NORM [1-6]. While concentration of the natural radionuclides in raw materials is usually low, but the concentration can be enhanced during manufacture processing [6-8] in some by-products. It is called TENORM (technically enhanced NORM) and the elevated radioactivity induces the increase of radiation dose. To protect people from the exposure, Korean government enforced the 'The Act on Protective Action Guidelines against Radiation in the Natural Environment' in 2012 and TENORM are under the regulation boundary to protect people from radiation exposure. One of the main concerns of the TENORM exposure is due to scrap iron pipes discharged in large quantities from the TENORM industries. Due to the diverse sizes of the scrap pipes, it is not easy to measure the radioactivity of the scales (Fig. 1). Titanium dioxide (TiO_2) processing factory is one of the NORM related industries in Korea. The factory treats titanium minerals and various feedstocks containing natural radionuclides. While the radioactivity in the raw materials is usually very low in the range of few Bq per gram, scales in the scrap pipes have elevated radioactivity during manufacturing processes [6]. In this study, we conducted in situ gamma spectrometry to the discharged scrap iron pipes from the factory. Action level (AL) is derived the in terms of dose rate by Monte Carlo simulation to estimate the radioactivity of the pipe scale. The derived AL can be applied to the inspection and assessment of discharged scrap iron from the NORM industries.

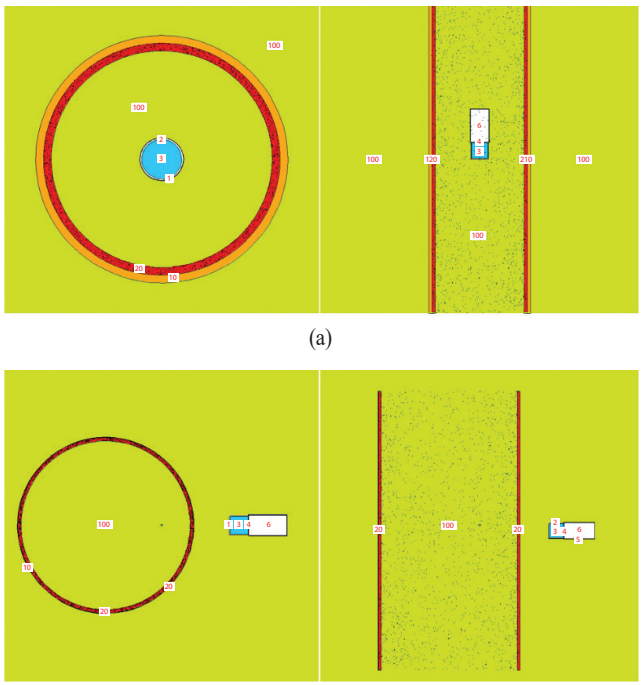
2. Materials and Methods

2.1 MCNP Simulation for Pipe Residues

MCNP is a Monte Carlo simulation code to conduct radiation transport. Before actual measurement, we conducted a simulation to estimate dose rate of the pipe by the detector as shown in Fig. 2. Internal diameter and thickness are considered as two shape parameters of the pipe which are able to affect the dose rate. In each shape, the thickness of scale was varied from 1 to 10 mm with uniform concentration. The length of pipe was fixed at 100 cm. Gamma radiations were included from isotopes such as ^{214}Pb, ^{214}Bi in the ^{238}U decay series and ^{228}Ac, ^{212}Pb, ^{212}Bi, and ^{208}Tl in the ^{232}Th decay series except below less than 1% emission probability. The density of the scale was assumed as 2.5 $g \cdot cm^{-3}$. The $LaBr_3(Ce)$ detector was placed either at the central position in the pipe or at the outside of the pipe with 10 cm away (Fig. 2). When the detector was placed inside, thickness of the pipe was fixed as 10 mm because the effect of the pipe thickness has little effect on the situation. Likewise, the diameter of pipe was determined as 30 cm because pipe diameter can be neglected when detector is positioned at outside. Table 1 shows the MCNP input conditions including detector location and other simulation conditions. At each condition, MCNP simulation was conducted to derive dose rate and energy spectra with F4 and F8 tally.

Table 1. Input parameters of the MCNP simulation for scale measurements

Input parameter		Content
Pipe	Material	SUS (density: 7.82 g·cm^{-3})
	Inner diameter	30, 50, 70, 100 cm
	Thickness	1, 2, 4, 6, 8, 100 mm
	length	100 cm
Scale	Material	$CaSO_4$ and $MgSO_4$ (density: 2.5 g·cm^{-3})
	Thickness	1, 2, 4, 6, 8, 10 mm
	Radionuclides	^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{212}Bi, ^{208}Tl
Detector		2”φ × 2” $LaBr_3$(Ce) scintillator



(a)

(b)

Fig. 2. Simulations of the scale at the inner surface of the pipe. MARK-B2 is located in (a) central position, (b) outside of the pipe.

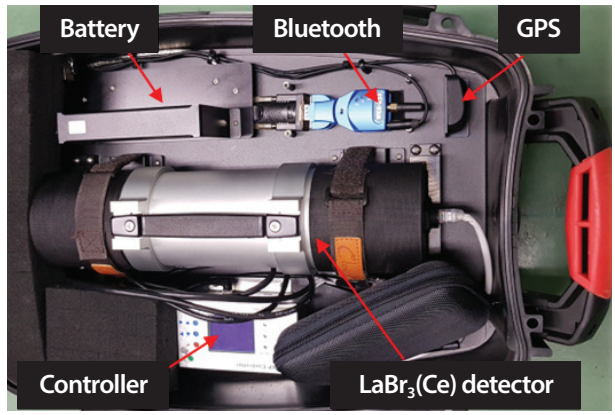


Fig. 3. The composition of the MARK-B2 detector system.

2.2 The $LaBr_3$(Ce) Detector System

The radiation detector system contains 2”φ × 2” $LaBr_3$(Ce) scintillation detector (51s51_B380, Saint Gobain, France) with a digital signal processing unit (SI Detection Company, Ltd., HAMPack MCA 527, Korea), controller, and GPS. The signal processing unit includes a high voltage supply, preamplifier, amplifier, and multichannel analyzer (MCA) with 1,024 channels. The $LaBr_3$(Ce) scintillation detector has high performance for gamma spectrometry with good energy resolution. But this type of scintillator has intrinsic background radiations from ^{138}La and ^{227}Ac, which exist in natural lanthanum element about 0.09% abundance. We applied a simple algorithm to subtract the intrinsic background radiation from the measured energy spectrum using the $LaBr_3$(Ce) detector [9-11]. The ambient dose rate can be calculated from the measured energy spectrum by applying dose rate-to-spectrum method (DRS method) [12]. The G-factor (nGy/hr/cps) which is the conversion factor between the count rate and the dose rate was derived from Monte Carlo calculation from the measured energy spectrum [9-12]. The equation (1) shows the assessment of the field gamma dose rate by subtracting the intrinsic background

(a) (b)

Fig. 4. In situ measurement of the scrap pipe with the MARK-B2 detector at (a) the central position and (b) the outside of the pipe.

radiation of the detector.

$$\dot{X} = \int n(E)G(E)dE - \dot{X}_{iBKG} \quad (1)$$

($\dot{X}$ = dose rate [nGy·hr^{-1}], $\dot{X}_{iBKG}$ = intrinsic background radiation)

2.3 Radiation Measurement of the Residual Scale of the Pipe

In situ radiation measurement was conducted to an iron scrap pipe discharged from the factory. The pipe was transported to a background location to prevent from the other radioactive residues. Fig. 4 shows the in-situ measurement conducted inside the pipeline. To position the detector deeply inside the pipe, a slider was inserted up to a depth of 10 cm, and measurements were taken. Repeating measurements at both the front and rear ends can enhance the accuracy of residual radioactivity concentration evaluations. The internal and external diameters of the pipeline were averaged at approximately 600 mm and 610 mm, respectively through repeated measurements. The length of the pipe was averaged to be around 1,000 mm and the pipe thickness was set at 3 mm, and the thickness of the scale was averaged as 2 mm.

3. Results and Discussion

3.1 MCNP Simulation Results

Figs. 5 and 6 show the dose rate from the MCNP simulation at each detector location. When the detector is positioned at the central position (Fig. 4(a)), it exhibited a precisely symmetric structure. As the internal diameter is increased, the dose rate exhibits an exponential decrease due to longer source to detector (STD) distance. For example, in the case of the pipe of internal diameter of 30 cm, the distance from the detector surface to the scale is approximately 10 cm. With the internal diameter of 100 cm, the distance from the detector surface to the scale is 45 cm. The increased diameter means the increased source volume, but the STD is more effective parameters in the central position. And the scale gets thicker, the dose rates go higher for increase of source amount. When the detector is positioned at the outside (Fig. 4(b)), as the pipe thickness is increased, the calculated dose rate decreases due to attenuation by the pipe. Considering these factors, to specify the effect of pipe thickness and diameter is necessary to estimate the amount of scale from outside of the pipe.

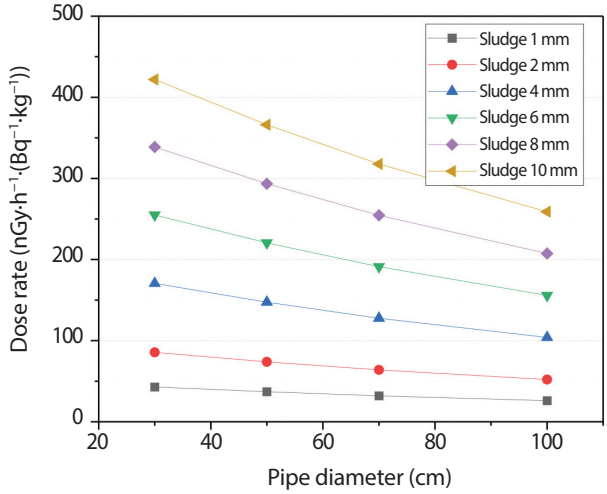


Fig. 5. Calculated dose rate for pipe diameter and sludge thickness in the central position of the pipe.

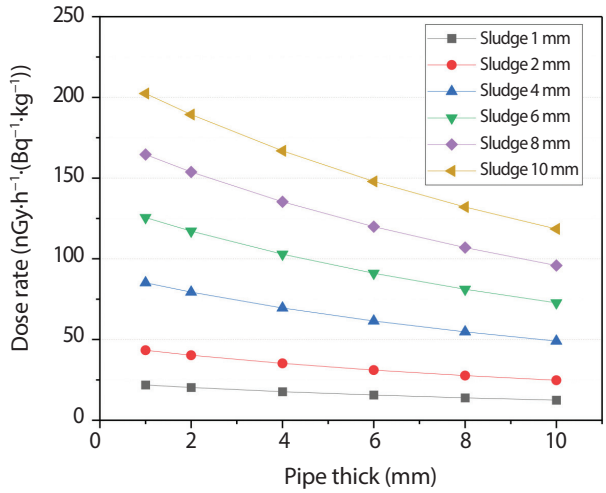


Fig. 6. Calculated dose rates for pipe and sludge thickness in the outside position of the pipe.

Table 2. Dose rate corresponding to action level which is referred as 1 Bq·g^{-1} for pipe scale

Scale thickness (mm)	Dose rate corresponding to action level (nGy·hr^{-1})									
	Central position				Outside position					
	Pipe diameter (cm)				Pipe thickness (mm)					
	30	50	70	100	1	2	4	6	8	10
1	43	37	32	26	22	20.4	17.7	15.62	13.88	12.42
2	85.6	73.8	64	52	43.4	40.4	35.2	31	27.6	24.8
4	170.6	147.4	127.6	104	85.4	79.4	69.6	61.4	54.6	49
6	254	220	191.2	155.8	125.8	117.4	102.8	91	81	72.6
8	338	294	254	208	165	154	135.4	119.8	106.8	95.8
10	422	366	318	258	202	189.6	166.8	147.8	131.8	118.2

3.2 Deriving Equivalent Dose Rate for Action Level

The action level is specified at 1 Bq·g^{-1} which is referred as registration criteria in 'The Act on Protective Action Guidelines against Radiation in the Natural Environment'. The equivalent dose rate corresponding to concentration of 1 Bq·g^{-1} was calculated (Table 2). When the inner diameter of the pipe is 30 cm, and the scale thickness is determined as 4 mm, the dose rate corresponding to the action level of 1 Bq·g^{-1} is 170.6 nGy·h^{-1}. Therefore, if the measured dose rate exceeds the sum of the background dose rate and action level, it is highly likely to exceed the action level. When the detector is located at outside, there is an overall decrease due to attenuation by the pipeline thickness. The simulation result for the scrap pipe measurements serves as a conceptual approach as one of the wide range of sizes and shapes of materials and equipment (M&E). In other words, developing conversion factors for radioactivity concentration for different measurement conditions is necessary. However, deriving conversion factors for such a diverse range of sizes and shapes of M&E involves extensive simulations and has

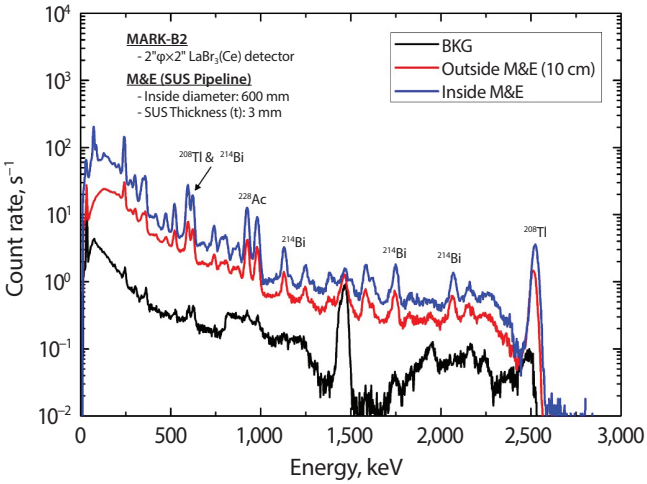


Fig. 7. Gamma spectrum measured at each point.

a significant drawback in that it requires repeated efforts whenever there are changes in size and shape. Therefore, it is necessary to create representative geometries for various types of industrial byproducts and to tabulate conversion factors for specific variables. Utilizing these results, it is desirable to determine dose rate corresponding to action levels for the target M&E, using a conservative approach.

3.3 In Situ Measurement of Pipe Scale

First, the background radiation was measured with the $LaBr_3(Ce)$ detector at 1 m height from the ground. The ambient gamma dose rate was measured to be 65.9 ± 5.3 $nGy \cdot h^{-1}$. Then, the detector was positioned inside the scrap iron pipe to measure dose rate. Fig. 7 compares the results of internal static measurements with external measurements. The average dose rate value at the central position was evaluated to be 3,011 ± 227 $nGy \cdot h^{-1}$ and it was estimated as approximately 92 $Bq \cdot g^{-1}$ in the scale.

4. Conclusion

To establish a design for radiation assessment for industrial by-products, we conducted an investigation into the current status of industrial by-products and regulatory frameworks. Based on this, we derived dose rate in unit activity in various size of pipe and derived action levels for on-site investigations of M&E for actual occurrences of industrial by-products in the facility. For the applicability assessment, we measured a scrap iron pipe occurred in a domestic TiO_2 processing facility. This application enabled the derivation of criteria for determining dose rate standards, which were subsequently applied to actual scrap iron pipes.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

REFERENCES

[1] International Atomic Energy Agency. Management of NORM Residues, IAEA Report, IAEA-TECDOC-1712 (2013).
[2] International Commission on Radiological Protection, Radiological Protection From Naturally Occurring Radioactive Material (NORM) in Industrial Processes, ICRP Publication 142, Ann. ICRP 48(4) (2019).
[3] International Atomic Energy Agency, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials, IAEA Safety Reports Series No. 49 (2006).
[4] European Commission, Practical Use of the Concept of Clearance and Exemption, Part II-Application of the Concepts of Exemption and Clearance to Natural Radiation Sources, Radiation Protection 122 (2001).
[5] International Atomic Energy Agency, Radiation Protection and NORM Residue Management in the Production of Rare Earths From Thorium Containing Minerals, IAEA Safety Reports Series No. 68 (2011).

[6] International Atomic Energy Agency, Radiation Protection and NORM Residue Management in the Titanium Dioxide and Related Industries, IAEA Safety Reports Series No.76 (2012).

[7] Y.Y. Ji, K.H. Chung, J.M. Lim, C.J. Kim, M. Jang, M.J. Kang, and S.T. Park, "Analytical Evaluation of Natural Radionuclides and Their Radioactive Equilibrium in Raw Materials and By-products", App. Radiat. Isot., 97, 1-7 (2015).

[8] Y.Y. Ji, C.J. Kim, J.M. Lim, H. Kim, and K.H. Chung, "Validation of the Quantification of Natural Radionuclides in Raw Materials and By-products Using Gamma-ray Spectrometry", Accredit. Qual., 21, 403-408 (2016).

[9] Y.Y. Ji, T. Lim, and W. Lee, "In Situ Gamma-ray Spectrometry Using an $LaBr_3$(Ce) Scintillation Detector", J. Radiat. Prot. Res., 43(3), 85-96 (2018).

[10] Y. Y. Ji, T. Lim, K. Hitomi, and T. Yajima, "Assessment of Radioactive Cesium Deposition Using Ground-based Gamma-ray Specrometry With a $LaBr_3$(Ce) Detector", J. Radiol. Prot., 39(4), 1006-1020 (2019).

[11] Y.Y. Ji, T. Lim, K. Hitomi, and T. Yajima, "Spectrometric Estimation of Dose Rate Induced From Radioactive Cesium in the Ground Using a Mobile Gamma-Ray Spectrometry Based on a $LaBr_3$ (Ce) Detector", Health Phys., 118(2), 215-225 (2020).

[12] Y.Y. Ji., C.J. Kim, K.S. Lim, W.N. Lee, H.S. Chang, and K.H. Chung, "A New Approach for the Determination of Dose Rate and Radioactivity for Detected Gamma Nuclides Using an Environmental Radiation Monitor Based on NaI(Tl) Detector", Health Phys., 113(4), 304-314 (2017).