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Primary study on holdup measurement of ²³⁵U in pipe using γ-ray spectrometry and Monte Carlo simulation

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Abstract The pipe holdup measurement is very important for decommissioning nuclear facilities and nuclear-material control and accounting. The absolute detection efficiencies (ε_{sp}) of full-energy γ rays peak under different source density distribution function have been simulated using the Monte Carlo (MC) software, and the counting rates (n_0) of the characteristic γ rays have been measured using the γ spectrometer followed by the calculation of the holdup. The holdup is affected by the energy of γ rays, distance at which they are detected, pipe material, thickness, and source distribution of pipe, especially source distribution at a short distance. The comparative test of ²³⁵U reference materials on the inner wall of Fe and Al pipes (the total mass of ²³⁵U is 44.6 mg and 222.8 mg, respectively) have been accomplished using this method. The determined result of ²³⁵U is 43.2 mg ($U_{0.95rel}$ =5.4%) and 216.2 mg ($U_{0.95rel}$ =3.2%), respectively, which are in accordance with the reference values.

Key words Holdup, Pipe, Source distribution, Monte Carlo simulation, γ spectroscopy

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1 Introduction

It is very important for decommissioning nuclear facilities, nuclear material control and accounting to identify and nondestructively assay radioactive material holdup truly in pipe of nuclear facilities. Although many researchers in the world focus highly on this measurement technology ^[1, 2], few published reports have been described in detail as yet.

2 Principle and method

The relationship between mass (*m*) of ²³⁵U measured by γ spectroscopy and the counting rates (*n*₀) of a certain characteristic, γ rays can be expressed by

$$m = \frac{Mn_0}{N_{\rm A} P \lambda \varepsilon_{\rm sp}} \tag{1}$$

where M = atomic weight of ²³⁵U; N_A = Avogadro constant; P = branching ratio of a certain characteristic

 γ ray of ²³⁵U; and λ = decay constant of ²³⁵U.

On the basis of the radioactive material decay theories, the holdup is calculated using $\varepsilon_{\rm sp}$ simulated by MC^[3], and n_0 is measured using HPGe γ -ray spectrometer. Since ε_{sp} is affected by γ -ray's energy, detection distance, material, thickness, and source distribution of the pipe, the corresponding ε_{sp} have been simulated using MC for different pipe materials, thicknesses, conjectural source density distribution functions, detection positions, and the energies. Among these influencing factors, the source density distribution is a key factor, although it is usually unknown. So, it is significant to develop means to discover the source density distribution of the pipe. On the one hand, the relation curves $\varepsilon_{sp}(d)$ (ε_{sp} vs. detection distance) and $\varepsilon_{sp}(v)$ (ε_{sp} vs. vertical displacement of the detector) have been obtained using MC under different conjectural source distribution functions. On the other hand, the relation curve $n_0(d)$ (n_0 vs. detec-

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tion distance) and $n_0(v)$ (n_0 vs. vertical displacement of the detector) have been obtained using γ spectrometer. The source distribution functions of the pipes can then be obtained by comparing $\varepsilon_{sp}(d)$ with $n_0(d)$ or $\varepsilon_{sp}(v)$ with $n_0(v)$. Thus, the pipe holdup (²³⁵U) has been measured primarily using the γ spectrometer and Monte Carlo (MC) simulation.

3 Design of simulation system

The simulation system (Fig. 1) consists of source, absorber, and detector. In the MC software, the center of the upper plane of the detection crystal is defined as zero point, the axis of horizontal pipe is defined as *X*-coordinate, and the axis of the cylindrical crystal is defined as *Z*-coordinate. And so, the source distribution functions for *X*- and *Z*-directions are defined as uniform, and the source distribution for *Y*-direction is defined as an exponential function under the practical conditions based on the aerosol theory. Due to gravitation, the quantity of 235 U at the bottom of the pipe is higher than that at the top of the quantity at the bottom to that at the top are defined.



Fig. 1 The simulation system.

4 Results

4.1 Effect of energy and thickness

Fig. 2 shows the $\varepsilon_{sp}(E)$ curves of the uniform distribution source in PVC, Al, and Fe pipes. Fig. 3 shows the $\varepsilon_{sp}(t)$ curves of different pipes with uniform distribution source.

In Fig. 2, for the pipes of uniform distribution, ε_{sp} rises rapidly at a low energy and decreases slowly with the increase in energy after the extreme points are reached. In Fig. 3, ε_{sp} varies with increasing thickness

of the material and decreases incoordinately according to the exponential law, and the more the atomic number of absorber materials, the more quickly ε_{sp} decreases.



Fig. 2 The $\varepsilon_{sp}(E)$ curves.



Fig. 3 The $\varepsilon_{sp}(t)$ curves.

4.2 Effect of detection distance

Fig. 4 shows the curves of ε_{sp} versus detection distance for uniform distribution source in PVC, Al, and Fe pipes.

In Fig. 4, for the pipes with uniform distribution, ε_{sp} decreases exponentially as the detection distance increases.



Fig. 4 Curves of ε_{sp} versus detection distance.

4.3 Effect of source distributions

Changing the source density distribution functions, ε_{sp} (²³⁵U:185.72 keV) for Al pipe has been simulated under measurement conditions. Figs. 5 and 6 show the $\varepsilon_{sp}(d)$ and $\varepsilon_{sp}(v)$ curves with different source density distribution functions, respectively. In Figs.5 and 6, "*y* uniform" denotes a uniform distribution for *Y*-direction. "*y* 2exp" denotes that the quantity of ²³⁵U at bottom of the pipe is twice than that at the top of the pipe for *Y*-direction. "*y* 1e *x* exp" means that the quantity of ²³⁵U at the bottom of the pipe is 1×10^{x} times that at the top of the pipe for *Y*-direction (*x*=1, 2, 3, 4, 8).



Fig. 5 The $\varepsilon_{sp}(d)$ curves of Al pipe.



Fig. 6 The $\varepsilon_{sp}(v)$ curves of Al pipe.

The $\varepsilon_{sp}(d)$ curves are very much related to the source distributions of the pipes. The shorter the distance, the stronger the relativity will be. The $\varepsilon_{sp}(v)$ curves are also very much related to the source distribution of the pipes.

The counting rate n_0 of the full-energy peaks of characteristic γ rays of ²³⁵U has been measured using the HPGe γ spectrometer, and $n_0(v)$ curve is thereby

obtained. The reason for the obvious shape difference between $\varepsilon_{sp}(v)$ and $n_0(v)$ curves is analyzed using a linear relationship between n_0 and ε_{sp} (Eq.(1)), so is the shape difference between $\varepsilon_{sp}(d)$ and $n_0(d)$ curves. By changing the source density distribution functions, the simulation causes the calculation results to gradually approach the experimental results, and the source distribution is finally achieved.

5 The results of measurement

The mass of ²³⁵U reference materials on the inner wall of the Fe and Al pipe (total mass of ²³⁵U is 44.6 mg and 222.8 mg, respectively) has been measured, respectively, using this method. Figs.7 and 8 show $n_0(d)$ and $n_0(v)$ curves for Al pipe.



Fig. 7 The $n_0(d)$ curve for Al pipe.



Fig. 8 The $n_0(v)$ curve for Al pipe.

Comparing Fig. 7 with Fig. 5, and Fig. 8 with Fig. 6, it was found that the source distribution function was about 10 times (corresponding to "y le l exp"), so ε_{sp} is obtained using MC simulation precisely under the measured conditions, and the determined results have been obtained (Table 1).

Pipe material	S.D.F. for Y-direction	\mathcal{E}_{sp}	n_0/s^{-1}	<i>m</i> _{U-235} /mg	U _{0.95rel} /%
Fe	Uniform	5.03×10^{-4}	4.97	216.2	3.2
Al	10 times exp	1.42×10^{-3}	2.74	43.2	5.4

 Table 1
 The results of measurement

6 Conclusions

The primary research results have shown that ε_{sp} is very much related to the source distributions of the pipe: the shorter the distance, the stronger the relativity. So, the source distribution of the pipe can be achieved primarily by comparing $\varepsilon_{sp}(d)$ with $n_0(d)$ curve, or by comparing $\varepsilon_{sp}(v)$ with $n_0(v)$ curve. Moreover, the pipe holdup can be accurately measured using nondestructive assay. The results of ²³⁵U for Al and Fe pipes are accordant with the reference values.

This research is yet not complete, and hence further work with regard to this research will be carried out.

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