Removal of uranium from gravel using soil washing method

Ilgook Kim*, Kye-Nam Kim, Seung-Soo Kim, Jong-Won Choi
Korea Atomic Energy Research Institute, 1045 Daedeokdaero, Yuseong-gu, Daejeon, 305-353, Korea
Corresponding author: ilgook@kaeri.re.kr

1. Introduction

The development of nuclear technology has led to increasing radioactive waste containing uranium being released and disposed in the nuclear sites. For example, South Korea has a large amount of soils contaminated with uranium which was generated during the operation of nuclear facilities. Fine grained soils with a size of less than 4 mm are normally decontaminated using soil washing and electro-kinetic technologies [1,2]. However, there have been few studies on the decontamination of gravels with a size of more than 4 mm. Therefore, it is necessary to study the decontamination of gravel contaminated with radionuclides. The main objective of the present study on soil washing was to define the optimal condition for acid treatment of uranium-polluted gravel.

2. Methods and Results

2.1 Preparation of Gravel

Gravel was obtained from the uranium conversion facility site in Korea Atomic Energy Research Institute (KAERI). The size and the uranium concentration of gravel were about 5.0 – 30.0 cm and 3.0 - 6.0 Bq/g, respectively. At first, the gravel was crushed to less than 30 mm in size using a jaw crusher. Then, the gravel pieces were grinded using a pulverizer to less than 2 mm.

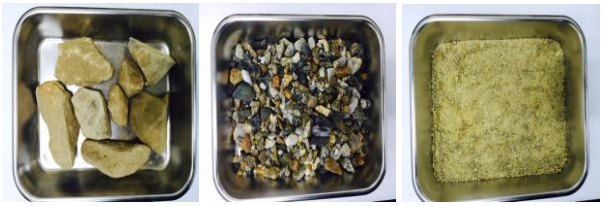

Fig. 1. Assortment of crushed gravel (initial state: left, after using a jaw crusher: middle, after using a pulverizer: right)

2.2 Soil washing

Soil washing was conducted in a plastic container with a total volume of 800 mL. For soil washing, 80 g of gravel and 160 mL sulfuric acid (H_2SO_4) solution were reacted using a shaking incubator at 60℃ and 150 rpm for 3 h. Sodium chlorate ($NaClO_3$) was also added as an oxidant to oxidize uranium compounds for improving the solubility of uranium in gravel [3].

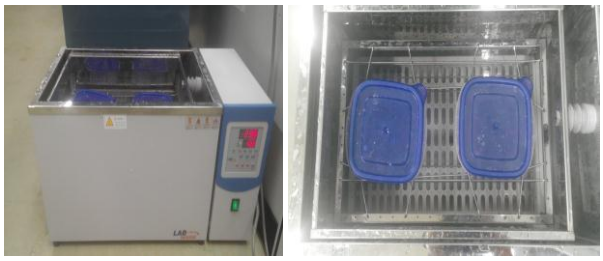

Fig. 2. Soil washing of U-contaminated gravel using a shaking incubator.

2.3 Measurement of radioactivity

IAEA has recommended the required natural uranium (^{238}U) concentration for self-disposal is below 1.0 Bq/g [4], and this value is also considered in the revised disposal regulation in South Korea. Thus, U-contaminated gravel should be treated to below 1.0 Bq/g.

The uranium concentration of gravel was analyzed using γ-spectrometry (Canberra, Genie 2000) by measuring the radioactivity of ^{234m} Pa (Energy 1,001 KeV). ^{238}U undergoes alpha-particle decay to daughter ^{234}Th (half- life = 24.1 days) to give secular equilibrium in less than 1 year, and ^{234}Th decays to ^{234m} Pa (half-life = 1.17 min) by beta-particle emission [5]. Since the gravel had been contaminated by natural uranium decades ago, the secular equilibrium among ^{238}U, ^{234}Th and ^{234m} Pa in this study has already been reached. The radioactivity of ^{238}U is corresponded to 45% of total radioactivity for natural uranium [6]. Accordingly, the clearance level of ^{238}U for self-disposal is 0.45 Bq/g.

2.4 Effect of particle size on uranium-removal efficiency

From the preliminary tests to obtain the proper particle size of gravel on washing condition, various sizes of gravel were treated to below the limit value for

self-disposal by washing twice with 1.0 M sulfuric acid at 150 rpm for 3 h. As the result of soil washing, gravel particles of above 2.0 mm and less than 0.45 mm could not reach the level of 0.45 Bq/g (Table 1). However, the size of 0.45~2.0 mm satisfied the self-disposal level indicating less than 0.30 Bq/g after 2nd washing. Therefore, we could conclude that the proper particle size of gravel for soil washing in removal of uranium was 0.45~2.0 mm.

Table I: Variation of uranium radioactivity according to particle size of gravel.

Particle size	Initial (Bq/g)	1st washing (Bq/g)	2nd washing (Bq/g)	Gravel recovery (%)
4.0 mm~	3.24	1.45	1.13	99.3
2.0~4.0 mm	3.76	1.48	0.67	97.0
1.0~2.0 mm	3.81	0.94	0.29	91.3
0.45~1.0 mm	3.94	0.96	0.30	82.5
0.075~0.45 mm	5.61	1.29	0.68	75.8
~0.075 mm	6.09	1.99	0.89	65.0

3. Conclusions

In this study, soil washing method was applied to remove uranium from gravel. The gravel was crushed and classified as particle sizes. The gravel particles were treated with sulfuric acid in a shaking incubator at 60℃ and 150 rpm for 3 h. The optimal particle size of gravel for soil washing in removal of uranium was between 0.45 and 2.0 mm.

REFERENCES

[1] K.N. Kim, U. R. Park, S. S. Kim, W. S. Kim, J. K. Moon, J. H. Hyun, Decontamination of gravels contaminated with uranium, Annals of Nuclear Energy, Vol. 72, p. 367-372, 2014.
[2] C. Kantar, B. D. Honeyman, Citric acid enhanced remediation of soils contaminated with uranium by soil flushing and soil washing, Journal of Environmental Engineering, Vol.132, p. 247-255, 2006.
[3] IAEA, Application of the concepts of exclusion, exemption and clearance, Safety Standards Series No. RS-G-1.7, Vienna, 2004.
[4] S. K. Bhargava, R. Ram, M. Pownceby, S. Grocott, B. Ring, J, Tardio, L. Jones, A review of acid leaching of uranite. Hydrometallurgy, Vol. 151, p.10-24, 2015.
[5] I. Adsley, A. L. Nichols, J. Toole, Decay of Th-234 and daughter 234m Pa in secular equilibrium: resolution of observed anomalies. DOE/CPR2/41/1/219, 1996.
[6] M. Gavrilescu, L. V. Pavel, I. Cretescu, Characterization and remediation of soils contaminated with uranium. Journal of Hazardous Materials, Vol. 163, p. 475-510. 2009.