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Removal of uranium from soil using full-sized washing electrokinetic separation equipment

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ABSTRACT

800 L washing-electrokinetic soil separation equipment was manufactured to decontaminate uranium contaminated soils from Korean nuclear facilities. For the removal of uranium at more than 95% from radioactive soil through soil washing-electrokinetic technology, several decontamination experiments were carried out. For a reduction of the generation of large quantities of metal oxides in a cathode, a pH controller is used to control the pH of the electrolyte solution between 0.5 and 1.0 for the formation of UO^{2+}. More than 60% metal oxides were removed through pre-washing, an electrolyte solution was circulated by a pump, and a metal oxide separator filtered the metal oxide particles. 60–65% of the uranium was removed from the soil by soil washing as a part of the pre-treatment. When the initial ^{238}U concentration of the soil was 22.2 Bq/g, the required electrokinetic separation time for the removal of below the clearance ^{238}U concentration of 0.4 Bq/g was 35 days, and the generated electrolyte solution rate was 1.4 ml/g. When the initial concentration of ^{238}U in the soil was higher, a longer decontamination time was needed, but the removal rate of ^{238}U from the soil was higher.

1. Introduction

A large number of nuclear facility sites have been contaminated by the leakage of radioactive waste solutions owing to corrosion of the waste solution tanks and connection pipes by their long-term operation around underground nuclear facilities. Therefore, a method to decontaminate a large volume of radioactive soil should be developed. Until now, a soil washing method has been studied to decontaminate soil contaminated with uranium, cobalt, cesium, and so on (Kim et al., 2007). However, it has a lower removal efficiency of nuclides from soils, generates a large volume of waste solution, and is impossible to apply to soil composed of fine particles. Meanwhile, the electrokinetic method has been recently studied as a new technology for soil decontamination, and has a higher removal efficiency of nuclides from soils but needs a longer decontamination time. Consequently, it is necessary to develop a washing- electrokinetic technology to remove uranium from lots of soil with a high removal efficiency during a short period, which first applies a washing technology for the removal of uranium above 80% from the soil, and then applies an electrokinetic technology for the removal of uranium above 95%.

As a process applied for the separation and extraction of contaminants such as heavy metals, radionuclides, or organic contaminants, electrokinetics was reported to be a feasible alternative for the decontamination of saturated or unsaturated soils, sludge, and sediments contaminated with uranium (both anionic: $(UO_2(CO_3)_2)^{2-}$ or cationic: (UO_2^{2+})) forms (USEPA, 1995; Cauwenberghe, 1997; Shrestha, 2004; Wallmann, 1994; Buck et al., 1996). Uranium contamination in native soil and at waste sites is present predominantly as uranyl (UO_2^{2+}) species, and therefore exists in an absorbed or precipitated form (Francis et al., 1993; Lindgren and Brady, 1997). Consequently, the effective mobilization of uranium cannot be accomplished without first solubilizing it. Complexation with organic or inorganic molecules to form positively charged uranyl complexes will result in desorption and solubilization of uranium and ultimate transport through a soil pore–water system under the influence of an electric field. Before uranium contaminant can be mobilized, it must first be converted into a soluble form. Complexation provides a method for solubilizing uranium. Electrokinetics provides an in situ method for mobilizing and removing a metal complex.

The main mechanisms of a contaminant's movement in an electrical field involved in electrokinetic technology are an electro-migration of the ionic species and electro-osmosis. Electro-migration probably contributes significantly to the removal of contaminants, especially at high concentrations of ionic contaminants and/or a

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high hydraulic soil permeability (Pamukcu and Wittle, 1992). The cathode reaction should be depolarized to avoid the generation of hydroxides and their transport in soil. The selected liquids, also known as purging solutions, should induce favorable pH conditions in soil, and/or interact with the incorporated heavy metals, so that these heavy metals are removed from the soil (Reddy et al., 2011). Recently, researchers have been investigating whether this method can be used to remove subsurface contaminants. They have also compiled published research on the use of electrokinetic techniques to decontaminate fine-grained soils, and have discussed some of the problems that occur during this process (Reddy and Chinthamreddy, 2003; Braud et al., 1998; Kim et al., 2001; Page and Page, 2002).

The separation of uranium (VI) and lanthanides was investigated by Macka et al. (1999), which applied capillary electrophoresis (CE) into the form of anionic complexes with arsenazo III in aqueous background electrolytes. They established that the performance depends strongly on the chemistry of the surface of the internal wall capillary. Also, the capillary electrophoresis and time-resolved laser-induced fluorescence (TRLIF) were found to be good methods to characterize the different complexes of uranium (Scapolan et al., 1997; Collins and Lu, 2001). Electro-osmosis is a dominant process where a direct current can generate an accelerated flow of water in the soil (Shapiro and Probstein, 1993). Electro-migration is the main mechanism for the electrochemical process when the contaminants are ionic or surface charged. It defines the movement of ions and ion complexes cross a porous media. The average mobility of the ions is about 10 times greater than that of electro-osmotic ability, but is superimposed or tied to the movement induced by Electro-osmosis, complementing each other (Lageman et al., 1989). In an electric field, the metal ions move toward the cathode by leaving the surface of the sediment. This process accelerates by the primary reactions of electrochemical processes through acidification of the sediment at the anode. The hydrogen ions can replace the metal ions from the sediment surface.

The washing method of this study, which belongs to the ex-suit method, extracts uranium from contaminated soil with a chemical agent (Man, 1999; Kuhlman and Greenfield, 1999). The DOE (Department of Energy) in the USA has studied a soil washing method for restoration of the Hanford and BNL (Brookhaven National Laboratory) sites (Fuhrmann et al., 1996; United States Department of Energy, 1995). Because contaminated soil has a specific size distribution, radionuclide sorts, and concentrations, it is necessary to develop a soil washing system suitable for its specific contamination characteristics. As a large quantity of waste solution is generated during soil washing (Robert, 1999), it is necessary to establish a recycling method for waste solution such as an ion exchange (Matejka and Zitkova, 1997; Agbenin et al., 1999), and chemical sedimentation (Tunay and Kabdasli, 1994; Kennedy, 1986) for a reduction of the waste solution volume.

In this study, 800 L washing-electrokinetic equipment suitable to the soil contamination characteristics of Korean nuclear facility sites was manufactured to decontaminate soil contaminated with uranium. To remove uranium at more than 60% from the soil through a washing technology, many soil washing experiments were carried out to obtain an optimum reagent type and its concentration. Also, in order to remove uranium at more than 95% from the washed soil, electrokinetic separation experiments using electrokinetic equipment of 800 L in size were carried out. The problems of the equipment were improved. The time required to decontaminate radioactive soil to under a clearance concentration level was yielded through demonstration experiments with the improved 800 L washing-electrokinetic equipment.

2. Materials and methods

2.1. Equipment manufacturing

Soil washing-electrokinetic decontamination equipment was manufactured to remove uranium from lots of soil with high removal efficiency during a short time, and consists of soil washing equipment, electrokinetic separation equipment, and waste solution treatment equipment for the removal of uranium. In addition, waste solution treatment equipment consists of precipitation equipment, concentration equipment, and a filter press. A schematic diagram of washing-electrokinetic separation equipment is shown in Fig. 1.

The soil decontamination process for the removal of uranium is shown in Fig. 2. 60–65% of the uranium was removed from the soil through the soil washing equipment, and the washed soil was then put in a soil cell in the electrokinetic separation equipment to remove the uranium at more than 95% from the soil. Meanwhile, the waste electrolytes released from the electrokinetic separation equipment, which have a low concentration of metal ions, are used for soil washing with the addition of nitric acid. Waste solution

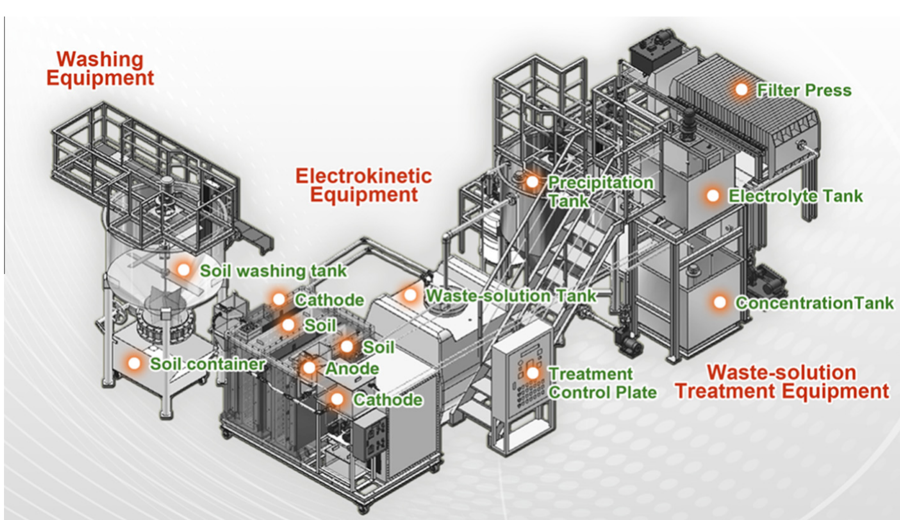

Fig. 1. A schematic diagram of the washing-electrokinetic separation equipment.