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Distribution characteristics of radionuclides in high-temperature melt phase during melt decontamination of decommissioned metal waste

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ABSTRACT

The purpose of this paper is to investigate radionuclides under various melting conditions that influence their distribution behavior during contaminated metal waste melting. Radioisotopes ^{60}Co, and ^{137}Cs were artificially used as the model contaminants. Most of the ^{60}Co remained in the ingot phase homogeneously and was barely present in the slag phase. The ^{137}Cs nuclides leave the melt caused by volatilization, and are essentially transferred to the slag and dust phase. They are then retained by filter systems. The captured ^{137}Cs in the slag phase is about 9% and 12% at a basicity of 0.82 and 1.42 respectively. The results of the demonstration test performed with a withdrawed steam generator from Young-Kwang Nuclear Power Plant shows that cobalt was distributed homogeneously on the ingot phase.

1. Introduction

Many nuclear facilities including NPPs will be shut down and decommissioned in the near future. A commercial nuclear power plant will produce 40,000 to 50,000 tons of metal waste during its decommissioning. Contaminated metallic waste represents a considerable storage volume as well as significant cost since it must be maintained and monitored indefinitely in secure storage. The high cost of either disposal or storage requires that the volume of the material be minimized. In Korea, research reactors (TRIGA MARK II, III) have been undergoing decommissioning since 1997. A large amount of radioactively contaminated metal waste is currently produced, and will continue to be generated during decommissioning and available decontamination. The decommissioning projects of the TRIGA MARK II and a uranium conversion plant (UCP) at KAERI have been completely finished. The decommission projects of TRIGA MARK III has been under way. Among the total 2200 tons of waste from the decommissioning of the TRIGA MARK II, such as concrete, soil, combustible and non combustible waste, more than 200 tons of radioactive metal waste was generated. In the case of UCP radioactive metal waste, approximately 200 tons was generated during the decommissioning project (Min et al., 2012). These decommissioning works resulted in various metal wastes such as stainless steel, carbon steel, aluminum, and copper. The total amount of carbon steel and stainless steel among the metal wastes from the nuclear facilities was up to 70–80% (Choi et al., 2004). Metal contamination generated from a nuclear facility can be of two forms, bulk or surface contamination. Bulk contamination usually arises from the neutron activation of nuclides during the service life of the components. Much of the metallic waste arising during decommissioning is only surface contaminated rather than activated. Surface activity can be loose contamination arising from the deposition of nuclides from the interfacing medium, i.e., an aqueous phase or gas phase during service. The deposited nuclides will depend on the environment of the component during service. Surface contamination can also be tightly bound, and this usually arises from the adsorption of deposited nuclides into the oxide layer formed on the metal. These require more aggressive decontamination techniques, such as melting, to remove them. The melt decontamination technology is the most effective treatment method for decommissioned metal waste. Melting for size reduction would require no prior surface decontamination and very little sorting of the waste material. In addition, the recycling or volume reduction of the metallic wastes through the melt decontamination technologies has merits from the view point of an increase in resource recycling as well as a decrease in the amount of waste to be disposed of resulting in a reduction of the disposal cost and an enhancement of the disposal safety (Buckley et al., 2004). Metal melting technology can be used to achieve three aims: a size or volume reduction of the waste, segregation or separation of the contaminants, and homogeneity of contaminants within the bulk metal. In addition, cost reductions may be possible because melting will create ingots in homogeneous waste form, which makes the waste characterization simpler and easier and stabilizes the final waste package. Therefore, there may not be a requirement for the additional packing of the waste. A proven melting technology is currently used for low-level

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waste (LLW) at several facilities worldwide (NEA, 1999; MSC, 2004; Socodel GROUPR EDF, 2012). These facilities use melting as a means of processing LLW for unrestricted release of the metal or for recycling within the nuclear sector.

In the present work, the behavior was studied for both cobalt and cesium in detail. The behavior of radioisotope ^{60}Co and ^{137}Cs among the molten ingot, slag, and dust phases have been investigated in an electric arc furnace.

2. Experimental procedure

Metal melting experiments were performed in one electric arc furnace (Fig. 1). The direct current (D.C) graphite arc melting system consists of one graphite electrode (I.D: 20 mm, L: 150 mm) and a copper crucible (I.D: 100 mm, H: 80 mm) which serves as a counter electrode. The cooling water flows in the surrounding area of the graphite arc electrode, copper crucible, and upper chamber. The input power can be supplied by controlling the D.C voltage from the D.C power supplier. Table 1 summarizes the melting experimental conditions. Inconel 600 material was used as the metal specimens at 10 mm × 10 mm × 1 mm. The metal specimens were contaminated with radionuclide nuclides such as ^{60}Co and ^{137}Cs. The contaminated metal specimen was dried in a vaccum oven at 60 °C. About an 800 g metal sample was placed into the graphite crucible with slag about 10 wt%. The turntable is pushed upright and an upper electrode makes contact with a bottom electrode on a turn table located at the bottom copper crucible; in addition, the arc is evolved. The gas is removed through an offgas treatment. Once the melting has been completed, molten metal is poured into the copper mold. The contaminated slag with radionuclides is easily separated from the ingot. Samples of metal, slag, and dust were examined for radioactivity. A cylindrical type sample was taken. The radioactivity of the sample was analyzed through a Multi Channel Analysis (MCA) using a high purity germanium detector. The slag formers were based on the constituents of silica (SiO_2), calcium oxide (CaO), and aluminum oxide (Al_2O_3). The slag bacisity is one of the most important parameters in a metal melting system. A slag is present on the top of the melt in a metallurgical furnace or vessel. The basicity ratio is defined as the ratio of basic to acidic oxides. The slag basicities used in this experimental were 0.82 and 1.42. The initial specific activity for a bacisity of 0.82 is ^{60}Co-4.07 Bq/g and ^{137}Cs-3.83 Bq/g, and for a basicity of 1.42 is ^{60}Co-3.15 Bq/g and ^{137}Cs-2.99 Bq/g. The various

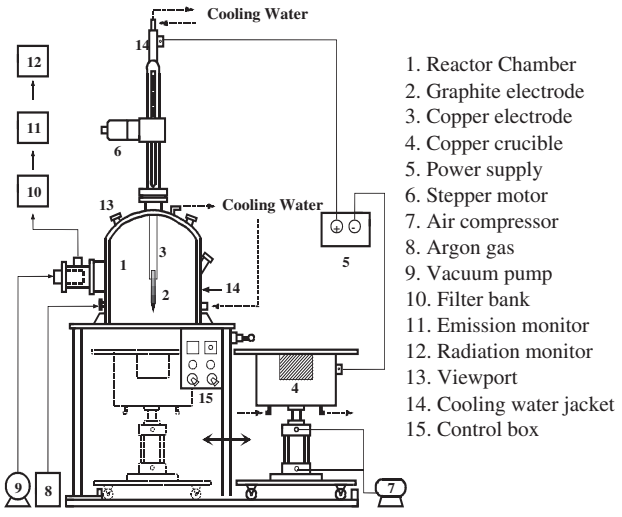

Fig. 1. A schematic of the electric graphite arc furnace.

stages of the melt decontamination process by an electric arc furnace are as shown in Fig. 2.

3. Results and discussion

3.1. Distribution

Melting processes operate above the melting point of the scrap metal so that decontamination reactions take place either between two liquid phases or between a liquid phase and a gas phase, making the reaction kinetics involved very rapid. The high temperature at which these processes operate also aids their kinetic efficiency. Fig. 3 shows the change in specific activity for ^{60}Co among the ingot phase, slag phase, and dust phase according to the slag basicity. The results of the melt decontamination show that the retention of cobalt in the slag is likely to be transient. For a slag basicity of 0.82, the initial specific activity is 4.07 Bq/g. Most of the cobalt remained in an ingot phase at 3.77 Bq/g and small amount of 0.15 Bq/g transferred to the slag phase. In the dust phase, cobalt nuclides were none detected. The change in the specific activity for cobalt nuclides for a slag basicity of 1.42 showed a similar tendency with a slag basicity of 0.82. Mostly, ^{60}Co remained in an ingot phase while a little bit of ^{60}Co transferred into this slag phase. The specific activity of the slag sample was found to be at 0.18 Bq/g. The radionuclide ^{60}Co is created by the neutron activation of ^{59}Co present in steel. All normal steels contain traces of cobalt, and steel that has been irradiated invariably contains ^{60}Co. In steel from nuclear installations, it may be present as ^{60}Co within the steel, or it may be as a surface contamination created by the deposition of ^{60}Co leached from elsewhere. Various studies for melt decontamination have been undertaken by researchers and workers (Gomer et al., 1985; Sappok, 1990; Nakamura and Fujiki, 1993; Larsen, 1985a,b; Pflugard et al., 1985; Min et al., 2008a,b; Schuster and Haas, 1990; Harvey, 1990). The conclusion regarding the melt decontamination was that ^{60}Co will almost be retained in the ingot phase in a uniform dilution in both an electric arc furnace and an induction furnace. The amount of ^{60}Co present in the slag is generally very low. Some was detected in the slag by some researchers when using an induction furnace (CEC Report, 1986). It appears that ^{60}Co can be transferred to the slag if it is initially present as surface contamination, and the melt is made under oxidizing conditions. Even under these circumstances, the retention of ^{60}Co by the slag is likely to be transient since the system in not in thermodynamic equilibrium. In addition, According to thermodynamic calculations such as a free energy calculation, the Ellinghan diagram (Ellingham, 1944), and the Kellogg diagram (Kellogg and Basu, 1960), the uranium can be completely removed from the ignore phase to the slag phase under equilibrium condition (Joanna, 1996; Anigstein et al., 2001; NeuschÜtz et al., 2005). According to test results of research paper, the optimum decontamination values was usually obtained near a basicity index of 1.5 (Abe et al., 1985; Uda et al., 1986; Joanna et al., 1996; Min et al., 2008a,b). But, another researcher reported that the optimum decontamination was achieved with a slag basicity index of 1.1. (Ren et al., 1994). The general methodology employed was to first consider the metal and slag reaction based on a thermodynamic calculation of Gibb's free energy to determine whether an element was likely to behave in the slag phase or ingot phase. Calculation of the free energy change for the reaction suggests that at 1600 °C the equilibrium distribution of cobalt between metal and slag will be approximately 800:1 as a minimum under strongly oxidizing conditions. The partitioning of a solute element between a molten metal and its slag under equilibrium conditions can be calculated form thermodynamic principles (Thurber and Mackinney, 1996). The calculated partition ratio of cobalt on slag phase based on