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Use of a single fuel containment material during pyroprocessing tests

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ABSTRACT

The use of a single stainless steel (STS) wire mesh basket as the fuel containment material for a series of pyroprocessing steps has been studied. The use of a single basket minimizes fuel loss and was enabled by transporting and using the basket containing the fuel from one test to the next without unloading it. The series of tests consisted of electrolytic reduction, LiCl distillation, electrorefining, LiCl–KCl distillation, and finally a second electrolytic reduction and a subsequent LiCl distillation step. While the electrolytic reduction of UO_2 was conducted in a LiCl–Li_2O molten salt electrolyte at 650 °C using the STS wire mesh basket as the cathode, the electrorefining was carried out in a LiCl–KCl–UCl_3 molten salt electrolyte at 500 °C, using the STS wire mesh basket as the anode. During the salt (LiCl and LiCl–KCl) distillation processes, the product of electrolytic reduction/electrorefining in the basket, which included metallic U and residual salts, was distilled at 850 °C under vacuum. The electrolytic reduction, electrorefining, and salt distillation processes were successfully demonstrated with the use of a single STS wire mesh basket through the entire cycle. However, an unstable intermetallic U–Fe layer was observed between the reduction product and the STS basket, when a cross section of the basket was studied after the salt distillation steps. The influence of the U–Fe layer on the electrolysis steps needs to be studied further in order to understand and quantify the lifetime of a single STS wire mesh basket during pyroprocessing.

1. Introduction

Developing sustainable nuclear fuel cycles that include the effective management of used nuclear fuel remains a formidable challenge. In this context, closed metal fuel cycles consisting of the use of metal fuel fast breeder reactors, pyroprocessing, and fuel fabrication by means of injection casting, have been developed with the goal of maximizing the efficiency of U usage and energy generation, while simultaneously minimizing radiotoxicity and waste generation, thereby increasing safety levels and economic efficiency. Pyroprocessing based on electrolysis using high temperature molten salts as the reaction medium, involves the reduction of the spent oxide fuel to its metallic form, through an electrolytic reduction process and the recovery of the fuel components by means of an electrorefining process (Benedict and McFarlane, 1998; Goff et al., 2011; Hur et al., 2008; Iizuka et al., 2008; Jeong et al., 2006; Kitawaki et al., 2008; Koyama et al., 2009; Laidler et al., 1997; Park et al., 2006; Serp et al., 2004; Simpson and Herrmann, 2008; Willit et al., 1992; Yoo et al., 2008).

The electrolytic reduction process is typically operated in a Li_2O–LiCl molten salt medium at 650 °C and utilizes a permeable basket carrying the spent fuel, which also acts as the cathode. The heat, volume, and radioactivity of the spent fuel loaded in the cathode basket can be reduced by the selective dissolution of its high heat-load fission products into the molten salt (Iizuka et al., 2008; Sakamura et al., 2006). When an electrical potential is applied between the cathode and the anode in the electrolytic cell, the actinide metal oxide is reduced to the metal at the cathode, whereas the O^{2-} ions, which pass through the cathode, are transported through the salt and discharged at the anode in the form of O_2 gas (Chen et al., 2000; Gourishankar et al., 2002; Herrmann et al., 2006; Hur et al., 2003, 2010; Redey and Gourishankar, 2003; Sakamura et al., 2008).

The metallic product obtained in the electrolytic reduction process is used as the feed for the subsequent electrorefining process. The superfluous salt (LiCl–Li_2O) present in the metallic product dissolves and diffuses into the bulk electrorefining salt (LiCl–KCl–UCl_3), which leads to a change in the eutectic composition of the LiCl–KCl–UCl_3 salt and the formation of UO_2 due to the small

Abbreviation: STS, stainless steel; OCP, open circuit potential; TGA, thermogravimetric analyzer; EDX, energy dispersive X-ray; ICP-AES, inductively coupled plasma-atomic emission spectrometry.

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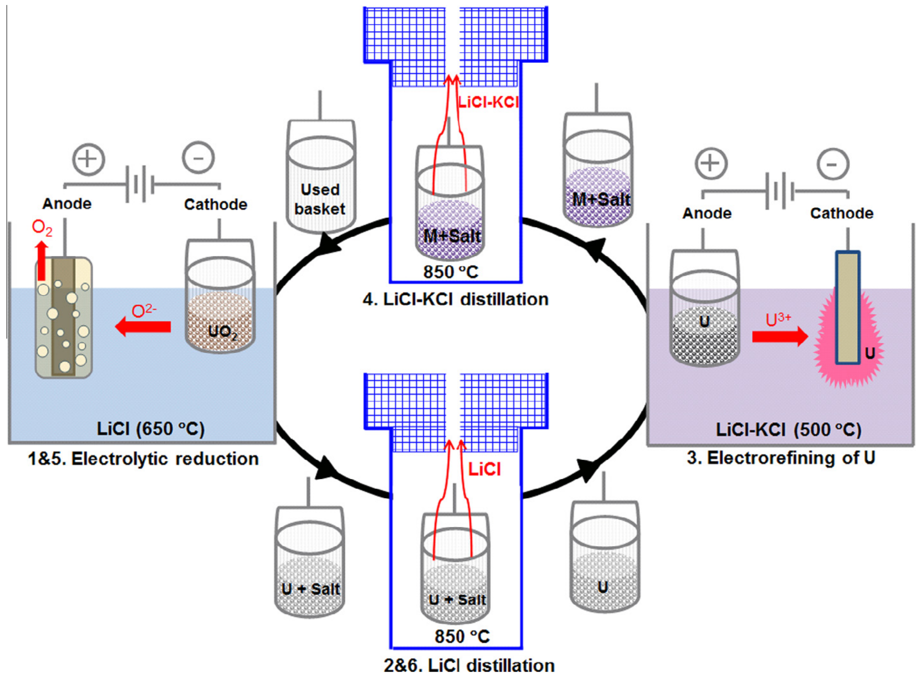

Fig. 1. Schematic diagram of the processes performed using the STS wire mesh basket: electrolytic reduction of UO_2 (Test1-OR), LiCl distillation of the reduction product of Test1-OR (Test2-LD), electrorefining of U (Test3-ER), LiCl–KCl distillation (Test4-LKD), electrolytic reduction of a fresh UO_2 in the used basket (Test5-OR), and LiCl distillation of the reduction product of Test5-OR (Test6-LD).

amount of Li_2O. Therefore, a salt distillation process is necessary to evaporate the superfluous LiCl from the metallic product, prior to the electrorefining process (Kim et al., 2013). In addition to salt distillation process, the metallic product obtained is typically transferred into a new permeable basket, which is then used as the anode for electrorefining, so that the eutectic composition of the salt is not affected.

In the electrorefining process, which is conducted at 500 °C, the actinide metals are electrochemically dissolved and transported to the bulk LiCl–KCl–UCl_3 molten salt via a permeable anode basket and simultaneously, high purity U metal is deposited on a solid stainless steel or graphite cathode ((Iizuka et al., 2009, 2010; Kato et al., 2006; Li et al., 2005; Vaven et al., 2008).

In general, a STS wire mesh sheet is used as the permeable basket material for containing the spent fuel, in the electrolytic reduction or electrorefining processes. The use of the STS wire mesh allows the molten salts to pass through with ease in addition to helping in decreasing the amount of residual salt in the fuel, a consequence of having sufficient number of openings in the mesh (Choi et al., 2012a, 2013, 2014; Herrmann et al., 2006; Herrmann and Li, 2010). In previous works involving the study of the pyroprocessing series, the metallic product produced during the electrolytic reduction process was unloaded from the STS wire mesh basket cathode and subsequently reloaded into a new basket, which was used as the anode during an electrorefining process (Herrmann and Li, 2010; Sakamura and Omori, 2010; Sakamura and Akagi, 2012). However, in practical applications, it is not possible to replace baskets when transitioning from one process to the next. Moreover, replacing baskets causes a hold-up of the fuel in the STS wire mesh basket, which can lead to a loss in the fuel and its utilization efficiency.

Keeping this in mind, in this work, we propose the use of a single STS wire mesh basket for a series of pyroprocessing tests. The present work aims at demonstrating the operation of each test in the series by using a single STS wire mesh basket and investigating the stability of the basket through the test cycle.

Table 1
Details of the series of tests performed in this study.

Order of the test	Type of tests	Test symbols
1	First electrolytic reduction	Test 1-OR
2	First LiCl distillation	Test 2-LD
3	Electrorefining	Test 3-ER
4	LiCl–KCl distillation	Test 4-LKD
5	Second electrolytic reduction	Test 5-OR
6	Second LiCl distillation	Test 6-LD

2. Experimental

2.1. Pyroprocessing test series

The series of pyroprocessing tests listed in Table 1 was performed by using a single STS wire mesh basket without unloading the fuel between process steps. The detailed procedure and a schematic illustration of the processes are presented in Fig. 1. First, the electrolytic reduction of the spent fuel in a LiCl (99% purity, Alfa Aesar)–Li_2O (99.5% purity, Alfa Aesar) molten salt medium at 650 °C was conducted by loading fresh UO_2 into a STS wire mesh basket (Test1-OR) and using it as the cathode. After the electrolytic reduction process, the basket was removed from the salt medium and directly transported and placed in an apparatus for salt distillation, where the residual salt in the basket was distilled at 850 °C under vacuum (Test2-LD). Next, electrorefining of the reduction product was conducted in the LiCl–KCl (99% purity, Alfa Aesar)–UCl_3 salt medium at 500 °C using the same basket as the anode (Test3-ER) and subsequently, the residual LiCl–KCl salt in the basket was distilled at 850 °C under vacuum (Test4-LKD). After the completion of one cycle of tests, fresh UO_2 was loaded into the same STS wire mesh basket used in the previous cycle and was employed as the cathode for the new electrolytic reduction step (Test5-OR), which was then subjected to a salt distillation process (Test6-LD).