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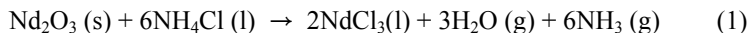
SYNTHESIS OF LANTHANIDE AND ACTINIDE TRICHLORIDES IN HIGH TEMPERATURE MOLTEN SALT

 S.-E. Bae^{1,2*}, H. Jeong¹ and T.-H. Park^{1,2}
¹*Korea Atomic Energy Research Institute, Daejeon, Korea*
²*University of Science and Technology, Daejeon, Korea*

*e-mail: sebae@kaeri.re.kr

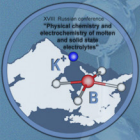
In a pyrochemical process, uranium dioxide, actinide dioxide, and lanthanide oxide have been converted to their trichloride forms for their dissolutions and electrodepositions. It is well-known that the lanthanide oxides such as Nd₂O₃, Gd₂O₃, etc. are not fully reduced to their metallic form during an electro-reduction process so that the lanthanide elements may not be dissolved during an electro-refining process. The lanthanide trichloride reagents are sold by a commercial reagent company whereas the actinide trichloride cannot be commercially purchased. However, in order to study chemical and electrochemical behaviors of the actinides in high temperature molten salt, it is essential to have the actinide trichlorides. In this work, in order to fully dissolve the lanthanide oxide into molten salt as well as to synthesize the actinide trichlorides in high temperature molten salt, we studied their direct dissolution and synthesis in the high temperature molten salt.

In order to test the conversion of the lanthanide oxide to the lanthanide trichloride in LiCl-KCl melt, we chose Nd₂O₃ and Gd₂O₃ as the representatives of lanthanide oxides. Fig. 1 shows a picture of Nd₂O₃ dissolved LiCl-KCl melt. We added Nd₂O₃ powder in transparent LiCl-KCl melt and shook the cell. The Nd₂O₃ was not dissolved and still existed as a form of colloids. The white blue of the picture is typical color of the Nd₂O₃ colloids in the melt. In order to dissolve Nd₂O₃ and get rid of the oxygen from the oxide, here we introduced ammonium chloride (NH₄Cl) in the melt. We expected that NH₄Cl may supply HCl in the melt. The HCl is very acidic molecule so that it can absorb the oxygen anion of the lanthanide oxides. Because the NH₄Cl is very volatile, we also expected that residuals of NH₄Cl after the reactions can be easily removed as a gas. The mechanism of the reaction can be described as shown below.



The products of the reaction are all gas phases except for the NdCl₃, which means that the by-product of the reaction can be easily removed from the melt.

Fig. 1b and 1c show the pictures of LiCl-KCl melt containing Nd₂O₃ after NH₄Cl was added. As soon as the addition of the NH₄Cl, there was a vigorous formation of gas bubbles and then the bubbles gradually disappeared in



5 minutes. After the reaction was completed, the melt turned into transparent and clean sky blue color (Fig.1c).

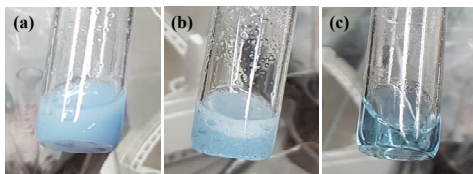


Figure 1. Pictures of Nd_2O_3 dissolved in LiCl-KCl melt at $450\text{ }^\circ\text{C}$. (a) Before and (b, c) after the addition of NH_4Cl .

Fig. 2 shows a cyclic voltammogram obtained from a W wire immersed in LiCl-KCl melt containing Nd_2O_3 after addition of NH_4Cl . It is obvious that cathodic and anodic currents around -2.0 V occurred. The current can be attributed to the electrodeposition and dissolution of Nd in LiCl-KCl melt. This result indicates that the NdCl_3 were directly formed from Nd_2O_3 with addition of NH_4Cl in the LiCl-KCl melt.

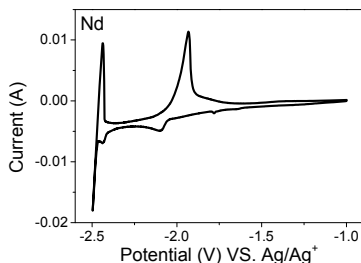


Figure 2. A cyclic voltammogram obtained from a W wire immersed in LiCl-KCl melt containing Nd_2O_3 after addition of NH_4Cl .

Actinide oxides have also been converted to their trichloride forms in the high temperature molten salt. The results will be given in the Conference.

References

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