Influence of the Rotating Speed on the Electrodeposition of ZrCl Metal with using a Rotating Cylinder Electrode

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1. Introduction

The dismantling technology of minimizing nuclear waste is gaining research interests as the facility become superannuated. In the case of Wolseong unit 2, the large amount of irradiated pressure pipes (Zr-2.5Nb) and calandria pipes (Zircaloy 2) which are corresponded to intermediate-level waste (ILW) due to including Nb-94, C-14, Ni-59, Ni-63 would be expected to generate. Thus, selective recovery of Zr metal from the alloy is required in order to achieve the low-, intermediated-level waste (LILW) as well as achieve reduced volume so that the waste can satisfy the acceptance criteria for radioactive waste disposal facilities. In this regard, the lab-scale experiments of Zr metal electrorefining using molten salt have been performed by various method [1-4].

This study aimed that investigation of hydrodynamic effect of RCE with respect to the rotating speed under the chloride based molten salt condition (LiCl-KCl-$ZrCl_4$, 500°C). The experiments were conducted to recover the ZrCl metal on the RCE surface when Zr ions are dissolved from Zr-2.5Nb metal. The numerical analysis was performed to back-up the experimental results from a phenomenological perspective. The numerical study was carried out by using COMSOL Multiphysics 6.0 [5]. The rotating speed were varied from 0 to 300, included laminar and turbulent regime. The largest *Re* is corresponded to 393. The critical *Re* is 200 [6].

2. Theoretical background

2.1 Recovery ZrCl metal

The conventional experimental studies have been tried to recover the Zr metal directly under molten salt system by applying a specific potential or current. However, the direct Zr metal electrodeposition showed disadvantage that is easily separated from the surface of electrode [4]. The present study adopted a novel approach method that Zr metal is recovered as the form of ZrCl. The deposited ZrCl metal on the electrode surface showed dendrite formation and it showed grown easily. Through the pyrolysis process, the recovered ZrCl will be converted to pure metal.

Figure 1 shows the cyclic voltammogram of LiCl-KCl-$ZrCl_4$ (1 wt%). The results present that Zr can be recovered as ZrCl when the R2 reduction peek potential (- 0.9V) applied [2].

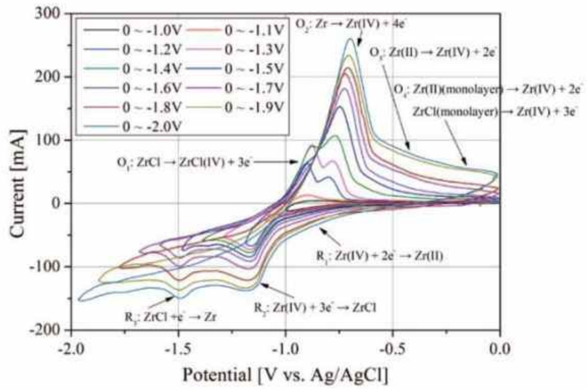


Fig. 1. Cyclic voltammogram of LiCl-KCl-$ZrCl_4$.

2.2 Rotating Cylinder Electrode

The RCE has been widely studied with regarding to the mass transfer and electrodeposition for a century. The advantage of the RCE is that it gains uniformly electrodeposited films resulting from the uniform potential and current at the surface of electrode and enhancing the mass transport with induced flow.

Eisenberg suggested the mass transport correlation (1) described as non-dimensional numbers [7]. *Sh* is the Sherwood number defined as the ratio of the convective mass transfer and diffusive mass transport (kd/D). k is the mass transfer coefficient, d is the diameter of rotating cylinder electrode and D is the diffusion coefficient. *Re* is the Reynolds number defined as the ratio of inertial force and viscous force (ud/v). u is the peripheral velocity of cylinder and v is the kinematic viscosity. *Sc* is the Schmidt number defined as the ratio of momentum diffusivity and mass diffusivity (v/D).

$$Sh = 0.079Re^{0.7}Sc^{0.356}. \tag{1}$$

However, increase in rotating speed can induce the Taylor vortices flow resulting from interaction between the rotor and the wall. Schlichting suggested modified Taylor number as shown below:

$$Ta = Re\left(\frac{R_2 - R_1}{R_1}\right)^{0.5}. \tag{2}$$

Taylor vortices develop when the value above the 41.3. These are presented as cellular or toroidal vortices within laminar regime. When *Ta* is over than 400, turbulent flow develops [8].

3. Results and Discussion

3.1. Experimental results

Figure 2 shows measured potential and current according to the electrode rotating speed. Each value is the averaged value that measured for 5 hours.

When the rotating speed lower than 100 rpm, where the regime corresponded to laminar regime, the current decreases as the rpm increases. Both the anode and cathode potential increase as the rpm increases. Increase in potential means that reduction and/or oxidation requires more driving forces to occur. Decrease in current means that the reaction on the electrode surface is reduced. When the rpm is 300, where the regime corresponded to turbulent regime, the reduction current presented the highest value, corresponded to -130 mA and the anode potential presented the lowest value, corresponded to - 0.66 V. It seems that it has become a good ionic mass transfer to induce reaction properly. As the only difference is that the rpm has changed, hydrodynamical approaches would be carried out to interpret this phenomenon.

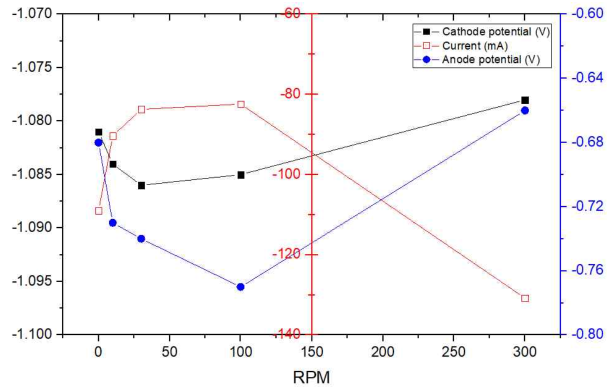


Fig. 2. Applied potential and measured current according to the RPM.

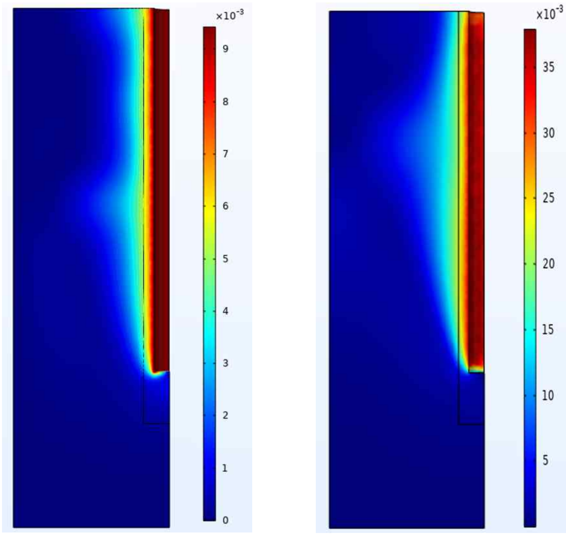
3.2. Numerical results

Monitoring the phenomenon such as ionic mass transfer and fluid flow pattern in high temperature molten salt is tough process. Thus, the numerical analysis was used to explain the experimental results. Table 1 shows the flow regime for the case of experiments.

Table 1. Flow regime for the experimental cases.

Case	rpm	*Re*	*Ta*	Regime
1	10	12.96	67.76	Vortices laminar
2	30	38.88	203.27	Vortices laminar
3	100	129.60	677.55	Turbulent
4	300	388.80	2032.68	Turbulent

Figure 3 shows the velocity contour for the laminar regime and turbulent regime. Fig. 3-(a) presents the laminar regime that corresponded to 30 rpm. The oscillated flow occurs at the vicinity of electrode. Fig. 3-(b) presents the turbulent regime that corresponded to 100 rpm. The oscillated flow become flatten and parabolic shape.



(a) (b)

Fig. 3. Velocity profile for the flow regime.

4. Summary and future work

The phenomenological study is conducted with numerical analysis in order to explain the experimental results. The results show the existence of hydrodynamic effect in the vicinity of the rotating vertical electrode.

For the laminar regime, the current decreases as the rotating speed increases. It seems that the vortices flow in the vicinity of electrode occurs inadequate mass transfer. However, for the turbulent regime, the current presents the highest value comparing to that of laminar regime. It seems that the influence of vortices flow become mitigated due to high rotating speed.

Because the influence of hydrodynamic boundary layer would be related to the diffusion boundary layer, the further study soon to be conducted with additional numerical analysis using electrodeposition model reflecting density gradient.

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