A Quantification Approach to Pyro-electrochemical Performance for Scale-up Design

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1. Introduction

Pyrochemical processing for the treatment of spent nuclear fuel consists of a series of molten-salt electrochemical reactors as the main processes. The pyrochemical process operated using a high-temperature molten-salt electrolyte is cost intensive since experiments are implemented under a radioactive environment.

A successful design of the electrochemical reactors is desirable for a product attainment at the planed throughput, at the projected performance and with the desired quality standards. Many trials of high cost experiment are not to predict the behavior of the commercial-sized equipment.

This is very important in the scaling of laboratory results to the engineering scale and then from engineering data to the commercial scale.

A larger scaled cell should be designed in favor of current/potential similarity to the corresponding planned electrochemical performance. It can be configured to get uniform current distribution on the electrode for scale-up.

Therefore, the perfect design and scale-up plays an important role in successful of this pyro-electrochemical technology for desired performance.

The topic of current distribution modeling is central to the analysis of electrochemical systems and has been addressed in a reference [1]. The prediction of current distribution is an essential step in the rational design and scale-up of electrochemical cells.

In this study, an approach to characterizing current/potential was proposed to quantify the electrochemical performance similarity. Focus is placed on discussing scaling parameters for the characterization of the current distribution.

2. Theoretical

For a given electrode configuration, the prediction of the current distribution is a complex problem and the difficulties further increase with increasing complexity of the cell geometry which is due to the different geometric and hydrodynamic conditions. It is possible to estimate certain objective trends, using a dimensionless group called Wagner number (Wa) given by

$$Wa = \frac{(d\eta/di)}{(1/\kappa)L} \quad (1)$$

where, (η/di) is the slope of the activation overpotential(η) to current density(i) dependence, κ is the conductivity of the electrolyte and L is a characteristic length. This number is the dimensionless ratio of the surface specific polarization resistance (η/di) and surface related ionic resistivity $(1/\kappa)L$, and is the relevant dimensional quantity determining current density distribution over electrode surface.

The Wagner number is used to characterize the relative importance of charge transfer control on the current distribution. The current distribution is expected to become more uniform as the Wagner number increases.

In addition, local Wagner number (Wa_x) can be derived throughout the electrode surface.

$$Wa_x = \left(\frac{\partial\eta}{\partial n}\right) / \left(\frac{\partial i}{\partial n}\right)\left(\frac{\kappa}{L}\right) \quad (2)$$

where, n represents the direction normal to the electrode surface. The average dimensionless Wagner number is estimated by integrating localized values over the electrode surface (S).

$$Wa_{average} = \frac{\int_S Wa_x dS}{S} \quad (3)$$

3. Results and discussion

Current and potential distributions are the most significant parameters characterizing the operation of the electrochemical cell. The current density on the electrodes is directly proportional to the reaction rate

and its distribution critically affects the refining quality.

Definition of the throwing power (*TP*) of a plating electrolyte has been used to characterize the uniformity of the thickness of the deposited metal in an electroplating system. An actual thickness of an electrodeposit obtained at different distances from the counter-electrode. Based on a semi-empirical correlation, if the Wagner number is relatively high, we expect very uniform deposit irrespective of the electrode geometry and cell scale (Fig. 1).

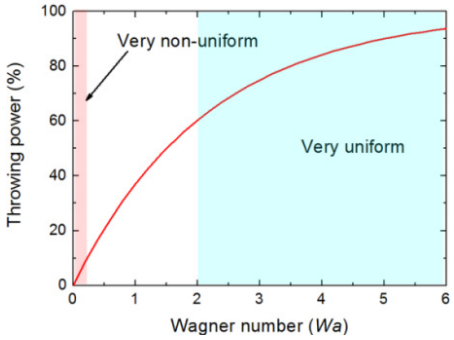

Fig. 1. Semi-empirical correlation between Wagner number and throwing power.

The Rotating Cylinder Hull (RCH) system [2] is optimized to be well suited for the study of plating processes by controlling mass-transport conditions. This system of the sophisticated design allows one to evaluate the effect of mass transport on current distribution. Theoretical expectation for the deposition rate at different distances from counter electrode is simulated by considering electrode polarization.

Copper sulphate electroplating baths are widely used in industrial applications to be informed of deposit quality standard. The current distribution on the RCH standard and corresponding quantification of deposit quality were determined. Dimensionless Wagner numbers based on cathode kinetics (activation and diffusion overpotential) were locally simulated on the rotating electrode as shown in Fig. 2.

This concept implies that for scale-up from the laboratory to the commercial scale the consequences of an enlarged size to current distribution have to be considered carefully in terms of the Wagner number.

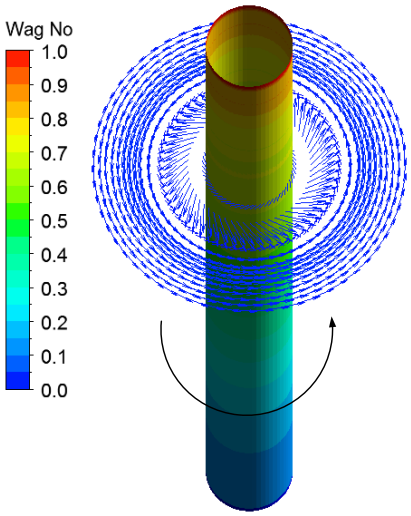

Fig. 2. Local Wagner number distribution along the rotating electrode (750 rpm).

4. Conclusion

This study shows that a theoretical approach to characterize current distribution on the cathode has been proposed for refining cell design. These means provide us design and operation guidelines to obtain a qualified deposit for specific applications.

REFERENCES

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