Computational fluid dynamics modelling of an electrorefiner for spent fuel partitioning

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Abstract

With renewed interest in spent fuel reprocessing in the US, pyroprocessing is being examined as a potentially proliferation-resistant reprocessing technology. This research presents a comprehensive computational fluid dynamics (CFD) modelling of an electrorefiner for pyroprocessing of spent fuel by taking advantage of the development of multi-physics analysis software aided by the latest computer technology. The modelling is based on using the multi-physics ANSYS software for a 3-D description of the processes and reactions involved. The 3-D CFD model simulates not only the fluid movements in the electrorefiner under the forced convection, but also the ion transfer under the applied current and a description of the electrochemical reactions involved. The model is applied to the Mark-IV electrorefiner design from the Idaho National Laboratory. An investigation of diffusion layer thickness under various operation scenarios was conducted and the results are presented herein. The use of the model to support design enhancements for pyro-processing is also discussed.

Introduction

Pyroprocessing is a non-aqueous method of reprocessing that employs high-temperature molten-salt based electrochemical technology. In the process, actinides are recovered as a group with no separation of pure plutonium contaminated with highly radioactive fission products. This presents advantages with respect to proliferation resistance compared to conventional aqueous reprocessing. The compactness of the technology allows on-site reprocessing, reducing the need for spent fuel transportation and the related potential security concerns. Additionally, electrochemical processing has the capability to treat a wide variety of spent fuel (metal fuel, oxide fuel, research reactor fuel) without imposing long cooling periods. The technology was developed at Argonne National Lab (ANL) [1-4] to recycle actinides from spent nuclear fuel and the development continued at both ANL and Idaho National Laboratory (INL) [5-11].

A central part of the pyroprocessing technology is the electrorefiner (ER) in which the actinides are extracted from the fission products. It is a steel vessel around 1 meter in diameter and 1 m high. At the bottom of the vessel is loaded a layer of cadmium liquid metal. The molten salt of LiCl-KCl is loaded on the top of cadmium pool. The spent fuel is chopped into small pieces and loaded into a steel basket. The basket is lowered into the molten salt region during the operation and can be used as anode. Two types of cathodes, the solid steel cathode and the liquid Cd cathode, are used to collect pure uranium or mixture of uranium, plutonium and other actinides. By applying electrical current and choosing different operation modes, spent fuel dissolves from the anode basket while uranium and other actinides are collected from the cathodes. The fission products are left in the salt and can be collected at later stage. The production rate and quality of the product depends on the system specifications, including the initial composition of the salt, the geometry of the anode and cathode, the speed of the rotating anode and cathode, and the applied electrical current and voltage. The performance of the electrorefiner varies significantly as a function of system configurations [3].

Various efforts have been made to model the ER in order to help improving the system design [1,5,12-15]. Most of them emphasise the modelling of the electrochemical reactions in the ER by using simplified geometry. In these modelling efforts, the details of the ER system geometry are typically not taken into account although the system geometry has a large impact on the distribution of species in the ER system. In particular, the concentration gradient in the very near region close to the electrode surface is determined by the system fluid dynamics directly affected by the system geometry. A recent work by the researchers at KAERI considered the impact of geometry by combing a 3-D computational fluid dynamic (CFD) model with a separate model (called REFIN) for the description of electrochemical reactions. In this work, the electrochemical reactions were described in one-dimensional manner while the fluid dynamics in the ER were treated with a three-dimensional CFD model [16]. The CFD model was used to determine the diffusion layer thickness which is one of the key input parameters to REFIN to update the electrical current density in the ER.

In this research work, development of a comprehensive computational fluid dynamics (CFD) model to simulate electrorefining operation for pyroprocessing of spent fuel is pursued. The work aims at a model that combines the system fluid dynamics with electrochemical reaction descriptions taking advantage of the development of multi-physics analysis software aided by the latest computer technology. The modelling is based on using the multi-physics ANSYS software for a 3-D description of the processes and reactions involved. The 3-D CFD model simulates not only the fluid movements in the electrorefiner under the forced convection, but also the ion transfers under the applied current along and a description of the electrochemical reactions involved. The model is applied to the Mark-IV electrorefiner design from the Idaho National Laboratory.

The current paper is a progress update of the research work with the focus on the investigation of diffusion layer thickness near the electrode surfaces under various operation scenarios.

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Project overview

This research work is intended to model the electrorefiner by implementing the embedded electro-hydrodynamics modelling ability in the commercial CFD software ANSYS CFX [17]. The goal is to simulate the impact on the ions transport from not only the fluid dynamic but also the applied electrical field. The electrochemical reactions are assumed to occur only at the interface between the anode or the cathode with the salt, or between the Cd pool with the salt. A quick equilibrium will be assumed at the interface considering the high operation temperatures in the ER. At this moment, the modelling is based on the Mark-IV ER design. This modelling exercise is to help to improve current ER design as well as to develop a general purpose modelling for pyroprocessing system innovation.

Two steps of modelling efforts are involved in this modelling project:

- *Step one*: A steady state 3-D electro-hydrodynamic CFD model is developed for the Mark-IV ER system. With the model, the electrical potential distribution, velocity field, diffusion layer thickness on the electrode surfaces can be investigated under various operating conditions. Diffusion layer thickness is one of the key modelling parameters representing the rate of material transfer through the electrode surface, which is used by other electrorefiner models [13]. In the current paper, investigation of the changes in the diffusion layer thickness under various operation scenarios is conducted and described.
- *Step two*: The electrochemical reactions at the interfaces between the electrodes and the salt system will be described to enable time-dependent mass transport analysis. The model for anodic dissolution will be included as part of this modelling step. The effect of liquid cadmium pool in the current ER design will also be modelled with the description of multi-elements in the system

Model development

ANSYS CFX is a commercial software widely used by researchers from different professions to simulate fluid behaviour in various engineering systems [17]. It has been successfully implemented to support the modelling of the ER for spent fuel pyroprocesing [16]. This project will take advantage of the enhanced electrohydrodynamic modelling ability in ANSYS CFX-12 will be taken for the purpose of developing general-purpose computer model development. As the diffusion layers between electrodes and the salt system play important role in the transport of ions in the ER, the detailed characteristics of diffusion layers will be captured in the model.

According to the published literatures [16], the thickness of diffusion layer is in the order of 1e-5 meter. This thin layer on the wall can be treated by the wall function in the ANSYS CFX with the use of fine grid; the thickness of the first element on the surface is less than 1e-6 meter. Because of its enhanced capability to treat the wall,effect the $k-\varpi$ turbulence model was selected instead of $k-\varepsilon$ turbulence model.

The molten salt was modelled as an incompressible viscous fluid mixed with uranium and the rest of the compositions in the salt. Multi-component flow model was adapted to simulate the movement of the fluid under the stirring of the anode basket and the cathode steel mandrel. The concentration distribution of uranium ion was obtained by solving the convection-diffusion transport equation:

$$\rho\frac{\partial Y}{\partial t}+\rho\nabla\cdot\left(\vec{u}Y\right)=\rho\nabla\cdot\left(D\nabla Y\right)$$

where ρ is fluid density, Y is mass fraction of uranium, $\vec{u}$ is the velocity of the fluid and D is the diffusion coefficient of uranium in the molten salt.

Uranium mass flux was assumed as a boundary condition at both the anode and the cathode surfaces, to represent the surface electrical-chemical reactions. The molten salt was represented

with the density of 1 551 kg/m^3, the dynamic viscosity of 0.00123 Ns/m^2, and the molar mass of 68.121 g/mol. The model simulated direct deposit operation: spent nuclear fuel is dissolved from the anode basket and uranium is deposited directly onto the cathode. A steady-state analysis was made to observe the distribution of uranium ions in the ER.

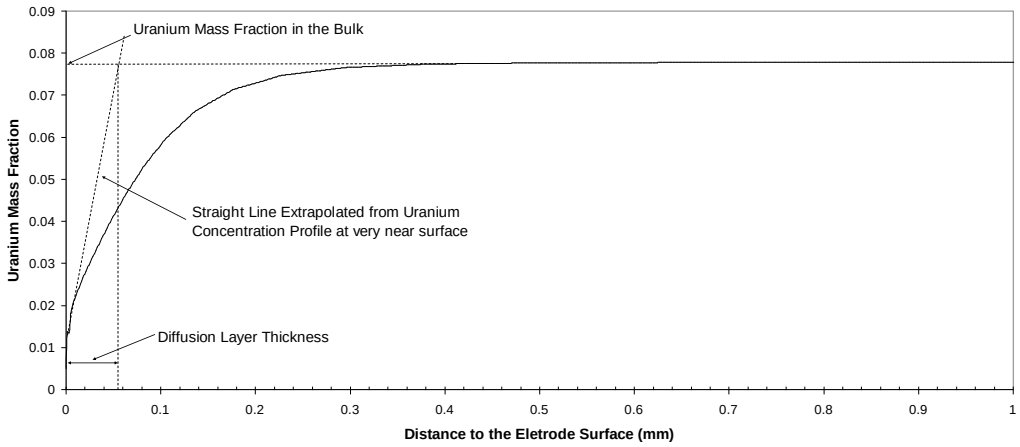
Scenarios

In this study, several operating scenarios were assumed to investigate the change of diffusion layer thicknesses. The base scenario assumed the operation with the anode velocity (ϖ_a) at 50 rpm, the cathode velocity (ϖ_c) at 5 rpm, the applied current (I) at 100 A, and the initial uranium concentration (wt_u) at 8 wt.%. In addition, four groups of scenarios (with a total of nine scenarios) were used to study the impact from the changes in the anode velocity, the cathode velocity, the applied current, and the initial wt.%, respectively:

- *Group 1*: Effect of cathode velocity (ϖ_a = 50 rpm, ϖ_c = 25, 15, 5 rpm, I = 100 A, wt_u =8%).
- *Group 2*: Effect of flux (ϖ_a = 50 rpm, ϖ_c = 5 rpm, I = 100, 150, 200 A, wt_u = 8%).
- *Group 3*: Effect of anode velocity (ϖ_a = 50, 75, 100 rpm, ϖ_c = 5 rpm, I = 100 A, wt_u = 8%).
- *Group 4*: Effect of initial wt.% (ϖ_a = 50 rpm, ϖ_c = 5 rpm, I = 100 A, wt_u = 6%, 8%, 10%).

The current efficiency, the ratio of current used for ion reduction to the applied current, was assumed at 100% for uranium ions. The uranium concentration in the salt was obtained after solving the mass transport equation in the ANSYS CFX. The diffusion layer thickness was determined by examining the concentration profiles near the electrode surfaces (Figure 1).

Figure 1: Determining diffusion layer thickness



Results of diffusion layer thickness investigation

The movement of molten salt is shown in the Figure 2 for the base scenario. The traces of the molten salt flow show that the salts are well mixed in the vessel. Figure 3 shows the distributions of the uranium concentration in the vessel. Three plane views of the ER are shown in the upper part of the figure. The concentration profile curves indicate that the uranium mass is for the most part uniformly distributed in the vessel except for the very small regions near the anode and cathode surfaces. The dissolved uranium from the anode generated higher concentrations at the surface which quickly drop due to the dilution by the movement of molten salt. The depletion of uranium due to electrical deposition on the cathode surface causes a sharp change of uranium concentration in the molten salt at the very near region close to the cathode surface.

Figure 2: Movement of molten salt in the ER

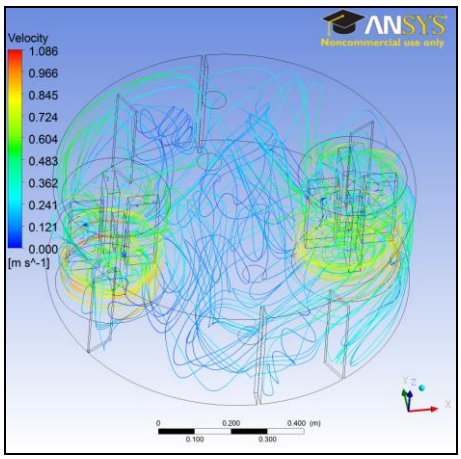
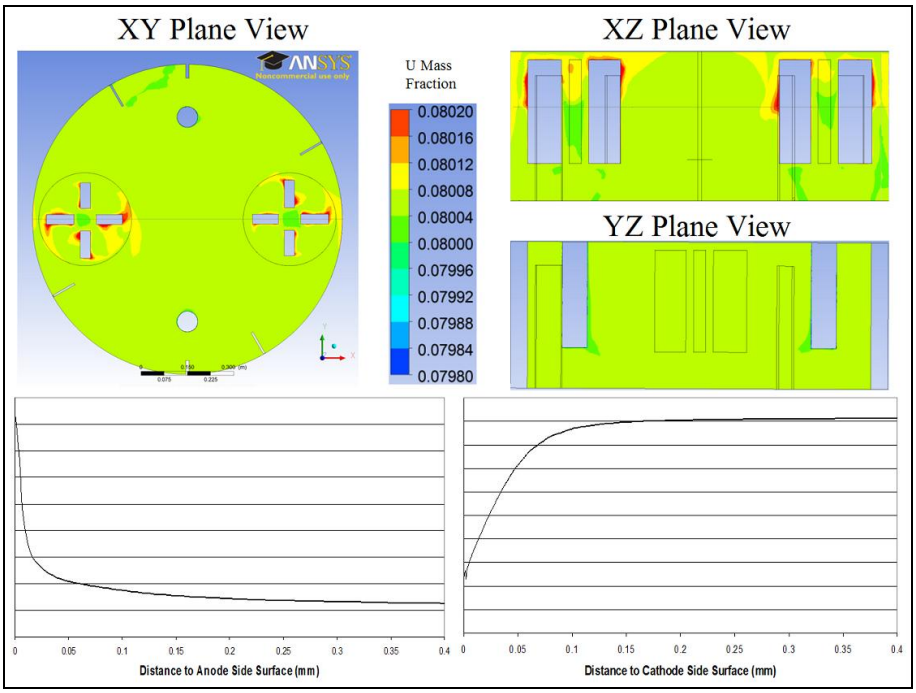


Figure 3: Distribution of uranium mass in the electrorefiner (in fraction)



The diffusion layer thickness was determined based on the predicted uranium concentration profiles near the electrode surfaces for all operating scenarios. The results are shown in Table 1 and Figure 4. The diffusion layer thicknesses were represented at four different regions near the electrode surfaces: i.e. at the anode bottom region, at the anode side region, at the cathode bottom region and at the cathode side region. The diffusion layer thickness was estimated to vary from 0.022 mm to 0.185 mm in these regions as being comparable to the published result (in the order of 0.01 mm). Except for the anode velocity, other parameters (such as the cathode velocity, the mass flux rate, and the initial uranium concentration in the molten salt) were found to have little impact on the diffusion layer thickness in these regions. The dominant effect of the anode velocity on the diffusion layer thickness appear to be due to the cross shape of the anode geometry, as its movement would strongly affect the molten salt velocity and the resulting mass distribution in the system. The diffusion layer thickness near the cathode bottom surface was shown to be relatively larger than in any other locations among all the simulated scenarios.

Table 1: Summary results of estimated diffusion layer thickness

Case*	Diffusion layer thickness at different location (mm)			
	Anode bottom	Anode side	Cathode bottom	Cathode side
Base case (c5,a50,wt8,X1)	0.025	0.032	0.167	0.027
c5,a75,wt8,X1	0.026	0.038	0.103	0.026
c5,a100,wt8,X1	0.031	0.037	0.092	0.022
c15,a75,wt8,X1	0.025	0.036	0.179	0.027
c25,a75,wt8,X1	0.025	0.034	0.185	0.028
c5,a75,wt6,X1	0.023	0.035	0.164	0.025
c5,a75,wt10,X1	0.025	0.027	0.172	0.026
c5,a75,wt8,X1.5	0.023	0.034	0.177	0.026
c5,a75,wt8,X2	0.024	0.033	0.174	0.026

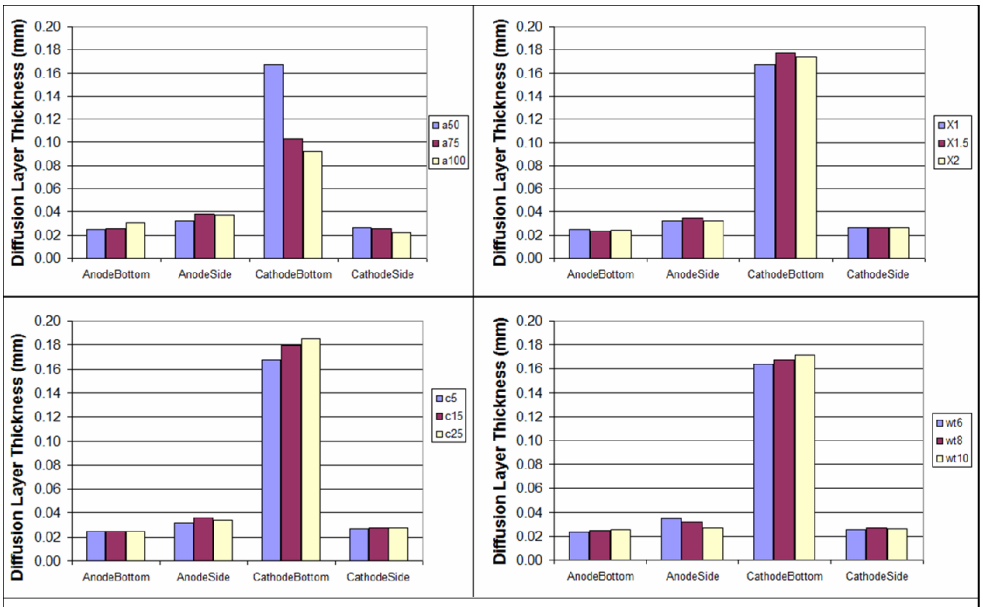
* a50, a75, a100 – anode velocity 50, 75, 100 rpm.

c5, c15, c25 – cathode velocity 5, 15, 25 rpm.

X1, X1.5, X2 – mass flux 1, 1.5, 2.0 times.

wt6, wt8, wt10 – initial mass fraction 6%, 8%, 10%.

Figure 4: Comparisons of estimated diffusion layer thicknesses under various operating scenarios



* a50, a75, a100 = anode velocity 50, 75, 100 rpm; c5, c15, c25 = cathode velocity 5, 15, 25 rpm
X1, X1.5, X2 = mass flux 1, 1.5, 2 times; wt6, wt8, wt10 = initial uranium mass fraction 6%, 8%, 10%

Discussion and future work

The current investigation showed that the diffusion layer thickness varies at different locations of the electrode surfaces and is affected the most by the anode velocity. The larger value of the diffusion layer thickness at the cathode bottom region would be due to the slower movement and inefficient mixing of the molten salt in the region. The results showed that increasing the anode velocity had a significant effect in reducing the diffusion layer thickness at the cathode bottom region. But increasing the anode velocity did not produce such a dramatic effect if the region was already well mixed. The observed large effect of anode velocity change would be due to the cross shape of the anode, which affects the salt movement more effectively than the cathode mandrel. Further study will be needed to determine the value of the optimal anode velocity.

In the actual operations of the electrorefiner, the deposits on the cathode will take the form of a porous metal as dendrites. This porous dentritic metal forms deposits on the cathode surface which will have a direct effect on mass transport. In this case, specifying a diffusion layer thickness near the electrode surfaces would be a complicated task. Future research will address the issue by developing supporting modelling capability to describe more complex shapes of the anode and the cathode.

The mass transport in the current study was assumed to occur only through diffusion in the diffusion layer. The migration flux driven by the electrical field was ignored due to its relatively small impact according to the published literature. Future work will include the investigation of the relative contribution to mass transport from both the migration flux and the diffusion flux by describing the time-dependent electrochemical reactions in the system.

As part of the modelling capability expansion effort, the model for anodic dissolution will be included and the effect of the liquid cadmium pool in the current ER design will also be modelled with the description of multi elements in the system.

The expanded modelling capability will lead into a general purpose modelling for pyroprocessing system innovation (e.g. a new ER design). The model will need to be validated by using experimental results and will be used to support design enhancements to fully explore the application of pyroprocessing technology for spent fuel reprocessing with enhancement in proliferation resistance.

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