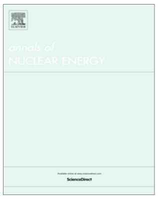
Annals of Nuclear Energy 112 (2018) 48–61

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene



Integrated model development for safeguarding pyroprocessing facility: Part II – Case studies and model integration

Wentao Zhou [a], Yafei Wang [b], Jinsuo Zhang [a,b,*]

[a] *Nuclear Engineering Program, Department of Mechanical and Aerospace Engineering, The Ohio State University, Columbus, OH 43210, United States*

[b] *Nuclear Engineering Program, Department of Mechanical Engineering, Virginia Polytechnic Institute and State University, Blacksburg, VA 24060, United States*

ARTICLE INFO

Article history:
Received 18 July 2017
Accepted 18 September 2017

Keywords:
Pyroprocessing
Electrorefining
Electrolysis
Model
Safeguards

ABSTRACT

In the part I, The developed model by the present study was validated by available experimental data. In the second part of the study, the major electrochemical processes of pyroprocessing, including electrorefining, electrolysis of actinide and rare earth were investigated by the model to study their separation performance under different operating conditions. This study provides insightful information about how one can alter the deposition by changing the operating conditions and also benefits the optimization of pyroprocessing design. An integrated safeguard model for the whole pyroprocessing was presented based on the electrochemical model. The integrated model shows how the material can be separated and tracked in the whole process of electrochemical separation.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

Due to a large amount of U and Pu involved in pyroprocessing, how to safeguard the facility has been a big challenge (Li et al., 2011) during its development. Basically, a facility can be mainly divided into three material balance areas (MBA), namely the head-end process (MBA 1), electrochemical separation process (MBA 2), and products as well as the waste storage area (MBA 3) (Ahn et al., 2012). The most difficult one to safeguard is the MBA 2 due to the accumulation of Pu and complex environment of an electrorefiner, inside which there are three major electrochemical processes: the electrorefining, actinide drawdown, and rare earth drawdown, as shown in Fig. 1 (Wang, 2017). The material flows include anode spent fuel charge, material inventory in electrolyte, U product, U/TRU product, and fission product waste. To determine the mass balance and detect any diversion scenario of nuclear materials, all these material flows need to be evaluated during the operation. A kinetic model development can be a good method for this purpose. Given the initial conditions, the material transport can be predicted, which can verify each other with other destructive assay (DA) or non-destructive assay (NDA) methods for the process monitoring. In part I, a robust model has been developed and validated by literature data. In the present paper, more cases about electrorefining, actinide drawdown, and rare earth drawdown were run to investigate their performance and determine the optimal conditions for the maximum separation between noble metals, actinides, and fission products. Eventually an integrated model simulating the practice electrochemical separation was studied to demonstrate its feasibility to capture each material flow.

2. Parameters identification

In this paper, all the simulations were run at the temperature of 773 K. For the model validations, only three elements Zr (IV), U(III), and Pu(III), were involved. But for the application of the model, totally 7 species were studied, such as Zr(IV), U(III), Pu(III), Am (II), Gd(III), Ce(III), and La(III). Because the potential of Zr is between U and other noble metals, all the noble metals will be retained in the anode if the dissolution of Zr can be avoided. Am is the actinide with a potential closest to the lanthanide species and Gd is the lanthanide with a potential closest to the actinides (Li and Simpson, 2002). Therefore, the separation performance of Zr, Am, and Gd can represent how the noble metals, actinides, and lanthanides are separated from each other. U and Pu are the actinides mostly concerned for safeguards and Ce and La represent other lanthanides. There are extensive studies about the activity coefficients and apparent potentials of elements in LiCl-KCl eutectic salt (Yamada et al., 2007; Zhou and Zhang, 2016; Zhang, 2014; Serp et al., 2006; Marsden and Pesic, 2011; Iizuka and Moriyama, 2010; Cumberland and Yim, 2014; Hoover, 2014; Zhou and Zhang, 2015, 2017; Caravaca et al., 2007). If multiple sources found

---

* Corresponding author at: Nuclear Engineering Program, Department of Mechanical and Aerospace Engineering, The Ohio State University, Columbus, OH 43210, United States.

E-mail address: zjinsuo5@vt.edu (J. Zhang).

https://doi.org/10.1016/j.anucene.2017.09.036
0306-4549/© 2017 Elsevier Ltd. All rights reserved.

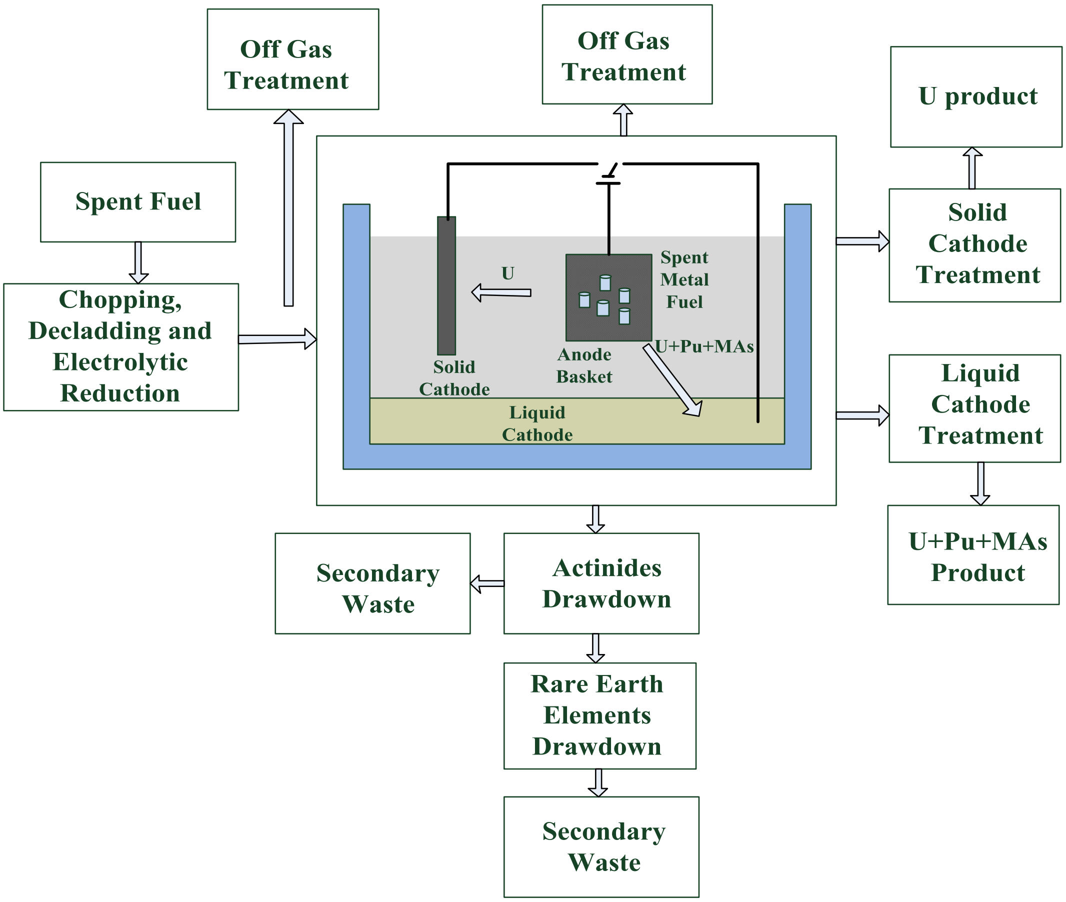


Fig. 1. The sketch flow of pyroprocessing (Wang, 2017).

in literature, data compilation by Zhang (2014) was used to get the average values. Here only the solubility of UCl_3, $PuCl_3$, and $ZrCl_4$ in LiCl-KCl eutectic salt was considered because concentrations of other species in electrolyte are too low to reach their solubility. The diffusion coefficients of element in porous Zr layer is related to the values in molten salt by

$$D_{porous} = k_d D_{ms} \tag{1}$$

The diffusion coefficients of U, Pu, Gd, and La have been reported in references (Zhang et al., 2015) and (Murakami and Koyama, 2011). However, no values were found for the Zr, Am, or Ce in liquid cadmium. Values of Zr and Am were taken as the same with U in the calculation and Ce was assumed to have the same values as Gd due to their similarity. The activity coefficients and solubility in Cd can be collected from reference (Zhang et al., 2015) and (Murakami and Kato, 2008), which are listed in Table 1.

Table 1
Solubility and the compounds formed when elements reach their solubility at 773 K, (except the data for Zr (Murakami and Kato, 2008), all the other data are from Zhang et al. (2015)).

Element	Activity coefficient	Solubility (at.%)	Compound
Zr	0.51	0.24 wt% (0.296 at.%)	Cd_2Zr ($ZrCd_2$)
U	89	1.11	U (UCd_0)
Pu	2.34×10^{-4}	1.86	$PuCd_6$
Am	8.00×10^{-5}	0.029	$AmCd_6$
Gd	4.40×10^{-7}	1.38	$GdCd_6$
Ce	9.19×10^{-9}	0.60	$CeCd_{11}$
La	3.36×10^{-9}	0.322	$LaCd_{11}$

The mass transfer coefficient of U in LiCl-KCl eutectic salt has been assessed in part I, the mass transfer coefficients of other species were derived from that of uranium via below equation (Gabe, 1974), which was applied to both molten salt and Cd.

$$K_i = \left(\frac{D_i}{D_U}\right)^{0.644} K_U \tag{2}$$

All the parameters used in the model are summarized in Table 2.

3. Application of electrochemical model

The part I indicates that the model is robust to predict the material transport and distribution in the electrorefiner. In this part, the model would be applied to electrorefining, actinide drawdown, and rare earth drawdown processes to investigate their separation performance under different conditions. Here all the anodes were solid used fuel anodes because they are the most ready-to-use form after the fuel is discharged from a fast reactor or obtained by reducing the fuel from a light-water reactor (LWR). Seven species, namely Zr, U, Pu, Am, Gd, Ce, and La were considered. Their weight percentages in used fuel are given in Table 3 in accordance to the composition of EBR-II used driver fuel (Li and Simpson, 2002). The Am was set to 0.5 wt% here.

3.1. Solid anode to solid cathode

The transport from the solid anode to solid cathode has been studied in Part I, which showed that Zr would be dissolved from

Download English Version:

https://daneshyari.com/en/article/5474748

Download Persian Version:

https://daneshyari.com/article/5474748

Daneshyari.com