A Preliminary Study on the Simulation of a Spent Oxide Fuel Metalization Process

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1. Introduction

A computer code simulating a metalization process of spent fuels is very useful from various aspects. There is the GC code[1] which is a computer code simulating such a situation but it is not available because its source has been unpublished. So it is necessary to develop a computer code that can simulate a metalization process of spent oxide fuels and finally, generate a database with information on the process.

In this study, we establish an algorithm through analyzing the properties of a metalization process of spent oxide fuels and develop a computer code to construct a database which provides overall information on the process. We also apply the code to the first batch.

2. Properties Analysis of the Metalization Process

The metalization technique is a method to reduce spent oxide fuels into metals and recover them. The method consists of four important sub-processes which are the reduction, electrorefining, transuranium extraction(TRUX), and salt recovery processes. In the reduction process, actinide and noble metal(NM) oxides are reduced into their corresponding metal in Li-LiCl molten salt at 650 ℃. Fission products(FP) and rare earth(RE) oxides are transformed into chlorides or mixed oxides. In the electrorefining process, the reduced actinide elements are transformed to trichlorides such as UCl_3 through reactions with $FeCl_2$. The remaining RE oxides react with UCl_3, which are then transformed to trichlorides, producing pure U and UO_2. In the TRUX process, pure actinides and RE elements are reduced from the trichlorides in a Cd-Li mixture. The salt recovery process transforms Li_2O to Li by a reaction with carbon.

3. Algorithm

The computer code has been developed based on PV-WAVE programming language. The code has composition elements of the spent oxide fuels, their initial quantities, free energies, and chemical reaction equations as input variables. After checking whether the chemical reactions occur with the standard reaction free energy, $\Delta G_r^{\circ} = \sum n \Delta G_f^{\circ}(product) - \sum n \Delta G_f^{\circ}(reactant)$, where ΔG_f° is a standard production free energy and n is a coefficient of each compound, the products or the remaining reactants and their quantities are determined. A database containing information on the process is finally generated.

The computer code is composed of four main modules, simulating the sub-processes which were explained earlier, and several other modules which handle the separation, delivery, and storage of the products and the remaining reactants. The four main modules generate the above chemical reactions, then carry out the following evaluations. The reduction module calculates 97.7 % of the quantities of the chlorides and mixed oxides, which are transferred to the electrorefining module along with the reduced metals. The remainder are transferred to the salt recovery module. The electrorefining module recovers the reduced NMs and calculates the

trichloride quantities and 98 % of the produced U quantities. The TRUX module evaluates the quantities of the pure actinides and the remainder. The CAT PROC module recovers only the U among the U, Li, and LiCl calculated from the TRUX module, and stores the Li and LiCl. The DIST CDLI module recovers the pure actinides transferred from the TURX module and stores the Cd and Li. The salt recovery module calculates 95 % of the Li quantities among the Li_2O obtained from the reduction module. The SP LI module stores the metalized Li along with the Eu and LiCl. The ZE SP module rejects 1 % of the quantities of the remaining elements in the salt recovery module, and then saves the remainder.

4. Comparison Analysis of the Results and the Database Construction

We have compared the results of our developed code with the previous code executed on the basis of the first batch. Table 1 shows only a few important products and their quantities, which are computed for some modules. Our results are almost the same as those of the previous code within the calculation error.

Table 1. The results of the developed code and the previous code.

Oxides	Initial Quantities	Reduction		TRUX and Salt recovery	
		Ref.[1]	This study	Ref.[1]	This study
UO_2	70.749	U : 70.749	U : 70.749	U : 69.989	U : 69.987
Li	350.0	Li (52.368,1.244) Li_2O(144.63,3.436)	Li (52.368,1.244) Li_2O(144.63,3.436)	Li : 297.97	Li : 297.97
LiCl	4500.0	(4394.3,104.37)	(4394.3,104.37)	0.651	0.651

The database produced as a result of executing our code, consists of many entries such as spent fuel elements, their initial quantities, free energies, and the products yielded from each module. This database will provide input data for the next stage of our simulation code.

5. Conclusions

We have developed a computer code to construct a database containing information associated with a metalization process of spent oxide fuels. As a result of the application of our developed code based on the first batch, it has shown that this code is reliable. It is possible to improve this code for a more detailed process than in this paper. We will further study the development of a computer code that could be applied to several batches based on this database.

REFERENCE

[1] RAhluwalia, and H.Geyer, The GC Computer Code for Flow Sheet Simulation of Pyrochemical Processing of Spent Nuclear Fuels, Nuclear Technology, v116, p.180-195(1996).