The identification of chemical form of fission products released in the operation of 100Mw chloride based Molten Salt Reactor(MSR) using FactSage modeling

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1. Introduction

The chemical form of fission product and its released amount during MSR operation provides a crucial information in the design of off-gas treatment system. In obtaining such information, chloride-based MSR is more disadvantageous than fluoride-based MSR already demonstrated by Molten Salt Reactor Experiment (MSRE). In this study, for the identification of chemical form of fission product and its released amount of chloride-based MSR, chemical thermodynamic approach[1] using Factsage coupled with extended MSTDB-TC database[2] necessarily for the phase description of chloride-based salts was applied.

2. Methodology

The methodology for the identification of chemical form follows the two steps approach as shown in Fig. 1. The first step is to calculate the total inventory of isotopes using neutronic code and the second step to predict the chemical form and the corresponding amount for isotopes considered through gibbs energy minimization approach such as FactSage. Since the basic database of FactSage is not sufficient in describing non-ideality of chloride-salt system, MSTDB-TC database was incorporated into FactSage as Private database.

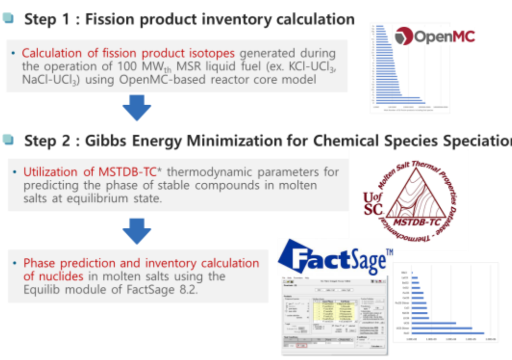

Fig. 1. Two-step based approach for the identification of chemical form of fission product

2.1 MSR model

For the evaluation of inventory of fission product, neutronics model based on OpenMC was employed. The chloride-salt fuel consists of 42.9 NaCl – 20.3 KCl – 36.8 UCl_3 fuel with 99% ^{37}Cl enrichment. The thermal power of the reactor was 100 Mwth and the volume fraction of Incore was assumed to 0.45. The number of particle was 50,000 with number of batch of 250 and number of inactive cycle of 100. The overall dimension of MSR model is schematically shown in Fig. 2. Using OpenMC code for chloride-based MSR, for 30 years of operation, we illustrated the calculated inventory of 25 fission product in Fig. 3. The major fission products were found to Zr, Pu, Mo, Xe, Nd, Cs.

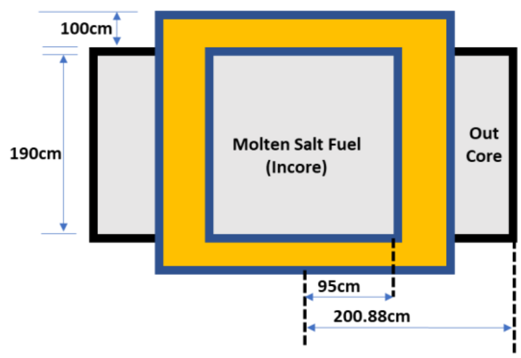

Fig. 2. Schematic diagram of chloride-based MSR.

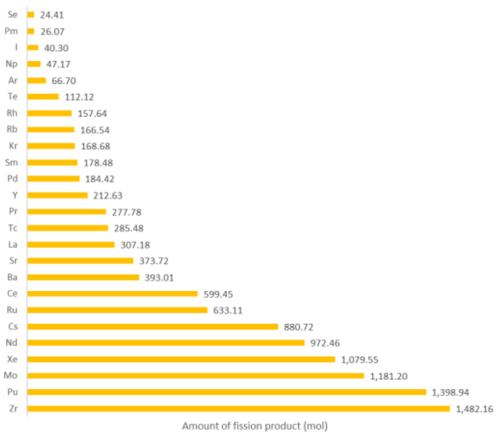

Fig. 3. Inventory of the generated fission products

2.2 Extended MSTDB-TC

Original MSTDB-TC covers various chloride-salt system including uranium chloride such as UCl_3 and UCl_4. However, in describing phase equilibrium of major fission products, some important systems were found to be neglected such as KCl-$ZrCl_4$. For the description of the chemical species in correct phase, the model parameter for following systems were regressed to the experimental data and added to original MSTDB-TC database: KCl-$ZrCl_4$, NaCl-YCl_3, KCl-YCl_3, NaCl-$SmCl_3$, KCl-$SmCl_3$, NaCl-$PrCl_3$, KCl-$PrCl_3$. The pure component physical properties of $PmCl_3$ were also included in the extended database from other physical database program and literature. The calculated phase diagram by the fitted model parameter were demonstrated in Fig. 4

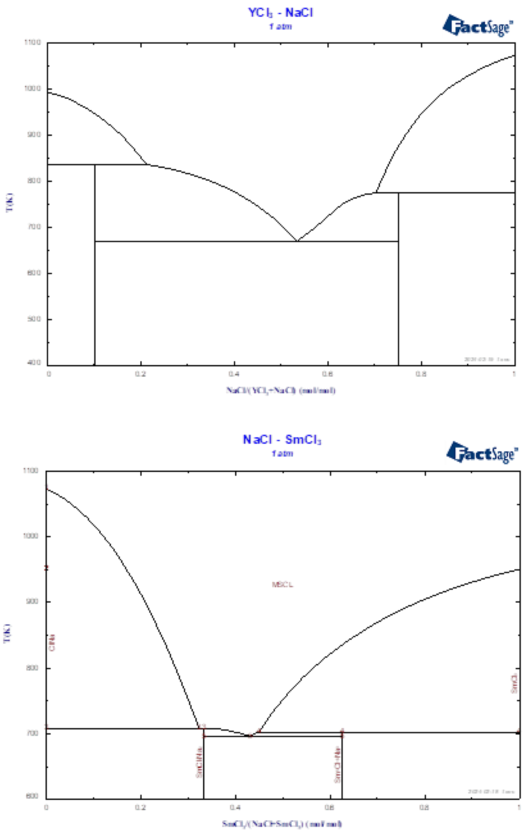

Fig. 4. The constructed phase diagram of YCl_3-NaCl and NaCl – $SmCl_3$ using the fitted parameter

2.3 Identification of chemical species of fission products

Equilib module of FactSage 8.2 allows to estimate the chemical species and the corresponding mass of fission product in element through gibbs energy minimization. Among all inventory, besides moisture and oxygen, 46 element species from Chloride to Americium were chosen and given to the input for Equilib with its mass. As required database, the extended MSTDB and FactPS were employed. In equilibrium modeling, metal composites such as URu_3, URh_3, UPd_3, Mo_5Ru_3, Mo_9Pd_{11} for solid phase and hydrogen gas and hydride such as H_2Se and H_2Te for gas phase were not considered. In Fig.5, major chemical species in gas, solid and liquid phase were demonstrated. During reactor operation of 30 years, except noble gases such as Xe, Kr and Ar, HCl, HI and hydride of volatile element, SnH_4 and SbH_3, were expected to be formed in gas phase and in solid phase, noble metals such as Mo, Ru, Tc, Pd, Rh and Uranium oxychloride due to the effect of moisture were found as major chemical species. In solution (liquid) phase, rare-earth chlorides including lanthanide were found, consistent to non-volatility of the chlorides.

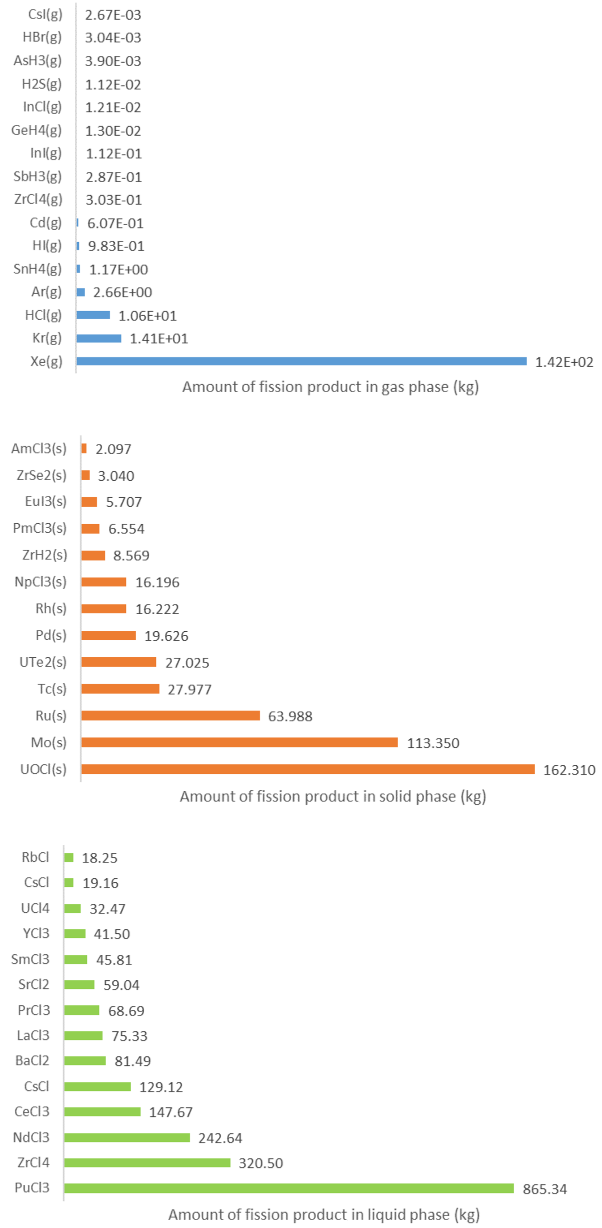

Fig. 4. The estimated chemical form and the distribution of fission products

3. Conclusions

In this study, using extended MSTDB-TC database and Equilib module of FactSage, the chemical form of fission products was preliminary identified and its distribution of were estimated. More careful evaluation for the assumptions employed is in progress to rationalize the methodology used in this study

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