Thermodynamic Databases Development in the System U-Zr-Ce-Cs-Fe-B-C-I-O-H and its Application in Simulating Phase Equilibria in Multicomponent Debris in Nuclear Accidents

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ABSTRACT

In case of severe nuclear accidents involving melting behavior of nuclear fuels at high temperatures, highly-volatilizing behavior of fission products (FP) would exert a significant degree of influence on radiative pollution and public exposure in surrounding areas.

In particular, it is of quite importance to accurately evaluate the volatizing behavior of cesium (Cs) and iodine (I) – bearing chemical species as notable FP.

However, in severe nuclear accidents, molten nuclear fuels (basically, UO_2-ZrO_2 system) would react with various kinds of surrounding materials in fuel container (e.g. Fe in stainless steel) and refractories (including B_4C) to form multicomponent debris.

Therefore, phase equilibria in multicomponent debris should be accurately evaluated for understanding FP volatizing behavior from debris.

Although thermodynamic computing package such as FactSage and thermodynamic databases have been developed for evaluating such complex phase equilibria in multicomponent systems even available for nuclear accidental situations, Cs, I-bearing systems have not been well treated yet.

In this study, we constructed the original thermodynamic databases in the system *U-Zr-Ce-Cs-Fe-B-C-I-O-H* featuring Cs, I-bearing systems, which consist of oxide, iodate and metal (including borides and carbides) databases.

A lot of available thermodynamic data were collected from literatures to be reproduced by calculations. Some complex intermediate oxide and iodate compounds including Cs were also taken account.

In addition, as a major volatile species, CsI thermodynamic property were optimized as a function of temperature based on the precise heat capacity measurements conducted by some of the authors.

We confirmed that the calculated phase diagrams by the present set of the databases well reproduce the corresponding literature data in various kinds of subsystems in the above multicomponent system.

Then, we attempted to simulate phase equilibria and volatizing behavior of Cs, I-including species in multicomponent debris under specific temperature and atmosphere conditions, where different methods and modules of calculating phase equilibria were adopted.

Finally, the reproducibility of the experimental results of phase equilibria in multicomponent debris by these simulations were discussed.