A Numerical Simulation of the Li Reduction Process for PWR Spent Fuel

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As a pyrochemical process, the Li process completely transforms spent fuels into their corresponding metals by using Li dissolved in molten LiCl at 650 ℃, except for the fission product and some rare earth elements. GC code[1] simulates the Li process which is composed of the Li reduction, Li recovery, electrorefining, and electrowinning processes. It is assumed that the code simulates a situation where the amount of Li is sufficient enough to completely reduce most of spent fuel oxides. Since the code is not available, so that the development of a similar code has been carried out[2-3]. This paper presents a computer code to simulate a process consisting of the Li reduction and Li recovery processes, and it compares its result with that of the GC code.

This developed program simulates chemical reactions to produce the quantities of products, which are calculated by SLG(simple but robust algorithm) by using a Newton-Raphson iteration as in the GC code. In the program, a database is established, which is composed of characteristical values related to the chemical reactions occurring in the Li reduction process, such as the gibbs free energy, activity coefficient, and the quantity of each compound. For simulating a real chemical reaction phenomenon, after determining the order of the chemical reaction of spent fuel oxides according to the magnitude of the gibbs free energy of each reaction given from the database, the amount of the products or the remaining reactants are calculated. Some data in the database have been assumed, only on the basis of the result of the GC code, because these values were not given in other references, which are some of the gibbs free energies and some of the activity coefficients of several compounds. When the activity coefficient was not given, 1.0 has been assumed except for $EuCl_2$, Li_3CeO_3, and Li_3NdO_3, whose activity coefficients have used the values presented in the database. In addition, a branching ratio, which is 1.0 in the case of a simple reaction, has been introduced for compounds having several composite reactions. In the other cases, the branching ratios of Eu_2O_3 and Sm_2O_3 have used the values assumed by regarding their composite reactions as only two reactions.

The performance test of the program has been carried out for 300 batches using a 20 kg batch of spent fuel. The quantities of the products produced from the Li reduction process for each batch have been accumulated. Of all the batch results, those of the 1st, 50th and 300th batches have been compared with those of the GC code and their relative errors have been calculated, which are the ratios of the quantities of each product calculated with this code and with the GC code, respectively. In the 1st batch, the two outcomes have been consistent with each other and, in the other two batches, they have been almost the same to within about a 6 % error except for Eu_2O_3 and Sm_2O_3 which had large relative errors for the 50th and 300th batches, respectively. Figure 1 shows the relative errors for the 50th and 300th batches. This developed program seems to be applicable to compounds, except for Eu_2O_3 and Sm_2O_3 having a large error. It is deduced that the errors resulted from their extremely simplified chemical reactions far away from the real reactions. It is required that the program be improved for the simulations of Eu_2O_3 and Sm_2O_3 through analyzing their reactions in detail. It is also necessary to add a user-friendly interface to the program.

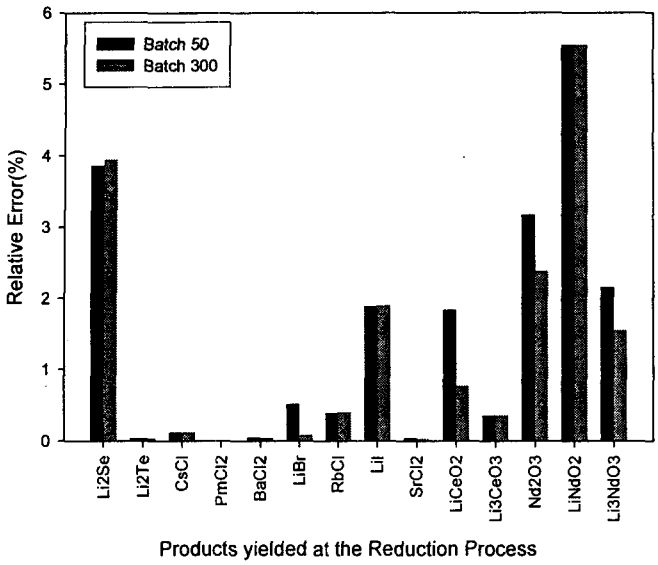

Figure 1. Relative Errors Comparison on the basis of the GC code result.

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