Chlorination of Uranium Oxides - containing Substances with Carbon Tetrachloride by Thermochemical Reaction

*Zhouran MA[1], Tadao YATO[2], Yoshiya HOMMA[2], Kenji KONASHI[2], Tatsuya SUZUKI[1]
[1]Nagaoka Univ. of Tech, [2] Tohoku Univ.

The dissolution of nuclear fuel debris is necessary for the accurate and precise analyses of nuclides. We have proposed the chemical conversion of insoluble nuclear fuel debris into soluble substances by thermochemical reaction. In the present work, the chlorination behaviors of UO_2, U_3O_8, simulated nuclear debris including UO_2 or U_3O_8 by themochemical reaction using CCl_4 were studied, and the converted substances were analyzed by XRD. We found that these substances can be chlorinated at 300℃.

Keywords: Actinides analysis, Uranium oxides, Nuclear debris, Chemical conversion, Pretreatment

1. Introduction

Fuel debris generated in severe nuclear accident (especially Fukushima Daiichi Nuclear Power Plants accident) contains nuclear fuel and core structural materials, and/or concrete materials. Thus, debris are difficult to dissolved into aqueous solutions. Analyses of actinides and several kinds of nuclides are required for planning of the effectual management of debris, and of the adequate decommissioning.[1]. Now we have proposed that these substitutes are chemically converted into easily soluble substitutes by thermochemical reactions.

2. Experiment method

The chlorination of uranium oxide and the simulated nuclear debris was carried out by mixing of these substances containing with uranium and CCl_4 in a Swagelok capsule made by 316ss. The components of simulated nuclear fuel debris are UO_2-ZrO_2-Fe. We investigated the chlorination of these substances by varying the mixing ratio of CCl_4, the heating time, and temperature(300℃and 400℃). Due to the different reactivity of triuranium octoxide and uranium dioxide, we studied the chlorination behavior of simulated nuclear fuel debris after oxidation.The samples were analyzed by XRD.

3. Results and Discussions

Color change of U3O8 samples after chlorination experiment was observed (Fig.1). The change in color may be due to a change part of the uranium,from tetravalence to hexavalent. In the cases of other samples, clear color changes were also observed. Both uranium oxides were confirmed to be converted into chloride, UCl_3 and UCl_4, by XRD analysis. Although the uranium oxides were remained. The experiments using simulated nuclear fuel debris, the products were black powder and a small piece formed by sintering because of prolong time heating. The XRD results showed that there was chlorides of uranium[2],[3], but there was still residual UO_2.

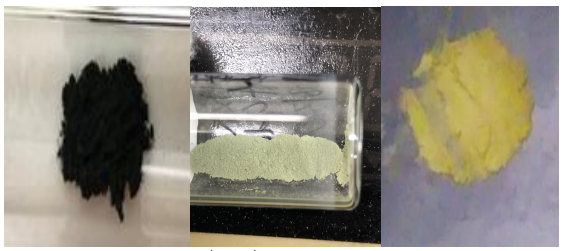

Fig.1 U_3O_8 powder(left),product after chlorination experiment (mid),product after prepared for XRD(right).

This work was partially supported by the Grant-in-Aid for Scientific Research (B) (KAKENHI No. 16H04628) and the Nuclear Energy Science & Technology and Human Resource Development Project (through concentrating wisdom) from the Japan Atomic Energy Agency / Collaborative Laboratories for Advanced Decommissioning Science.

References

[1] Washiya T., et al. Proc. 23th Inter. Conf. Nucl. Eng. p. 37
[2] Andresen A.F, Acta Crystallographica, 11, (1958)612-614
[3] Rudel S S , Kraus F . Dalton Transactions, 2017, 46(2):5835-5842.