1-9 Investigation of Compounds in the PuO_2-SiO_2 System
－Behavior of Si Impurity Contained in MOX Fuel－

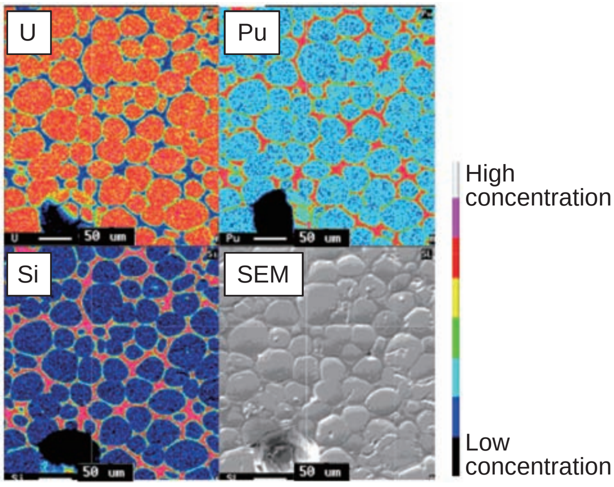

Fig.1-23 Element mapping images of MOX-6.9 wt% SiO_2 analyzed by EPMA
The cross-sectional surface of annealed pellets was analyzed by EPMA. The red area indicates a high concentration area of each element. Pu and Si were highly concentrated in the same areas and spread like network. Microstructural observation by SEM is shown in the lower right.

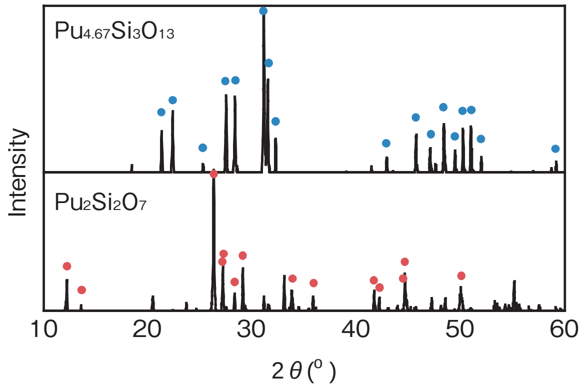

Fig.1-24 X-ray diffraction patterns of PuO_2-SiO_2 mixed powders after annealing
Specimens were analyzed by X-ray diffraction analysis. The $Pu_{4.67}Si_3O_{13}$ phase was precipitated in more conditions than $Pu_2Si_2O_7$. $Pu_2Si_2O_7$ was observed for the first time in this study.

Uranium and plutonium mixed oxide (MOX) fuels have been developed as fast reactor fuel. MOX fuels are fabricated from uranium dioxide (UO_2) and MOX powders by a mechanical blending method. The mixed powder is pelletized and sintered. In the mixing process, there is a possibility that the MOX powder is contaminated with silicon (Si), which is used as part of the ball mill pot. Therefore, it is necessary to evaluate the behavior of Si impurity. In this study, phase states in MOX-SiO_2 and PuO_2-SiO_2 systems were investigated to evaluate the behavior of Si in MOX fuels.

Specimens were prepared by mixing 6.9 wt% SiO_2 powders with MOX, and pelletizing and sintering at 2400 ℃. Fig.1-23 shows a cross-sectional mapping image of a pellet annealed at a low atmosphere (3.0×10^{-7} Pa) oxygen partial pressure (Po_2). Because Pu and Si are enriched in the same area along grain boundaries, Si can be considered to have reacted with Pu and formed compounds.

PuO_2-SiO_2 reaction examinations were carried out to investigate the precipitation conditions and chemical forms of the compounds. Specimens were prepared by mixing powders of PuO_2 and SiO_2 in molar ratios of 3:1, 3:2 and 3:3, and were then annealed as a function of temperature (1350~1700 ℃) and Po_2 (10^{-7}~10^{-10} Pa). X-ray diffraction patterns of the annealed specimens are shown in Fig.1-24. We observed that two kinds of compounds, $Pu_{4.67}Si_3O_{13}$ and $Pu_2Si_2O_7$, were formed as a function of temperature and Po_2. The phase containing $Pu_2Si_2O_7$ was precipitated in the specimens having mixing ratios of 3:2 and 3:3, and precipitation of $Pu_2Si_2O_7$ was limited to the region above 1600 ℃ and a Po_2 below 10^{-7} Pa. However, the $Pu_{4.67}Si_3O_{13}$ phase was precipitated in more conditions in comparison to the $Pu_2Si_2O_7$ phase. In addition to these two regions, there were conditions in which PuO_2 and SiO_2 compounds were not observed. These results show that, as a function of temperature and Po_2, Si impurity in MOX forms three kinds of chemicals: SiO_2, $Pu_{4.67}Si_3O_{13}$ and $Pu_2Si_2O_7$.

Thus, precipitation conditions for each compound were confirmed by investigating the behavior of Si impurity in MOX fuel. The acceptable level of Si impurity in MOX fuel is less than 1400 ppm, and the maximum amount of Si compound precipitated in MOX pellets is estimated to be less than 1 wt%. Therefore, Si impurity is considered to have a small effect on the properties of the fuel.

Reference

Uchida, T. et al., Phase States in the Pu-Si-O Ternary System, IOP Conference Series; Materials Science and Engineering, vol.9, 2010, p.012004-1－012004-5.