## 論文の内容の要旨

論文題目 Effects of Solute Elements in Irradiation Embrittlement of Reactor Pressure Vessel during Long-term Operation

(高経年化した原子炉圧力容器照射脆化における溶質原

子の影響)

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The neutron irradiation embrittlement of reactor pressure vessel steels is an important ageing issue for the safe long-term operation of light water reactors. Structural integrity of the reactor pressure vessel must be demonstrated throughout the entire operating life for normal operation and postulated accident events, notably pressurized thermal shock. This requires accurate prediction of reactor pressure vessel embrittlement based on the understanding of physical mechanisms considering various parameters, including material variables and irradiation conditions.

Solute clusters, composed of Cu, Ni, Mn, and Si, are one dominant microstructural feature formed under irradiation which cause hardening and accompanying embrittlement of reactor pressure vessel. The main objective of present study is to understand effects of solute elements, including Ni, Mn, and Si, in solute clusters of reactor pressure vessel steels under the conditions of long-term operation. The long-term operation condition for reactor pressure vessel could be represented using neutron irradiation following thermal ageing. Considering the interactions of Ni-Mn, Si-Cu, and Si-Ni, the research objectives are categorized as: to study effects of Ni and Mn in Cu-rich clusters during neutron irradiation following thermal ageing; to study effect of Si in Cu-rich clusters during neutron irradiation following thermal ageing; to study effect of Si on Ni-rich clusters at high dose. The change of solute effects from thermal ageing condition to neutron irradiation condition is also investigated and the role of solutes in understanding reactor pressure vessel embrittlement during long-term

operation is discussed.

The model alloys Fe-Cu, Fe-Cu-Ni, Fe-Cu-Ni-Mn, and Fe-Cu-Si were prepared for neutron irradiation following thermal ageing experiments. Additionally, high dose ion irradiation experiment was carried out on the model alloy Fe-Ni-Si. For the alloys, the microstructural analysis was conducted using atom probe tomography. The principal results obtained are as follows:

- (1) Mn is found to be critical for the formation of solute clusters. The addition of Mn to Fe-Cu-Ni reduced the ageing time required to achieve a similar microstructure of Cu-rich clusters from 225 to 90 h. This indicates that Mn can accelerate solute diffusion through thermal vacancy, thus accelerating kinetic process of solute clustering. Mn significantly increased the number density of Cu-rich clusters during the neutron irradiation even though the consumption amount of matrix Cu was almost unchanged. It is suggested that besides reducing the formation energy of Cu nuclei, Mn may also enhance the heterogeneous nucleation of Cu because of its strong interaction with self-interstitial atoms.
- (2) A strong thermodynamic relationship between Ni and Mn is found. In the alloys Fe-Cu-Ni-Mn, an increase of Ni alloying content enhanced the Mn concentration in the clusters as well as the Ni concentration. Moreover, for the alloys Fe-Cu-Ni-Mn with different Ni alloying contents, in the clusters the concentrations of Ni and Mn exhibited 1:1 relationship. The 1:1 Ni-Mn concentration relationship in the clusters remained unchanged under the neutron irradiation. These results indicate that 1:1 Ni-Mn binding can be energetically stable, even in the irradiation environment. It can be a driving force for enhanced concentrations of Ni and Mn in the clusters.
- (3) Si atoms hardly cluster with Cu, but largely cluster with Ni. Si may suppress coarsening of Cu clusters under thermal ageing. Except this, evidence for Si to have effect on Cu clusters was not detected. Moreover, the Si concentration in the clusters in Fe-Cu-Si was very low, indicating that Si atoms hardly cluster with Cu. However, Si should interact with Ni to form high number density of Ni-Si clusters in Fe-Ni-Si.

The present study contributes to ageing management and nuclear safety by increasing the basic knowledge of reactor pressure vessel embrittlement. Notably, this study found that the dominant role of solutes can change as the relative contributions between thermal vacancies and irradiation produced point defects are changed. A change in neutron flux can alter the relative contributions between thermal vacancies and irradiation produced point defects. Therefore, the disparity in neutron flux is important for the analysis of reactor pressure vessel data from different sources, e.g. boiling water reactor irradiation, pressurized water reactor irradiation, or material testing reactor irradiation. Even in particular reactors, the neutron flux on the inner surface of the reactor pressure vessel could be considerably different from that at the location of the tested specimens in a surveillance capsule. Suggestions for future studies are also provided based on the present study.