Overview of irradiation stability of oxide particles in ODS steels for nuclear systems

Xiaodong Mao, Ho Jin Ryu*, Min Ho Kim, Sung Soo Kim, Jinsung Jang
Nuclear Materials Research Division, Korea Atomic Energy Research Institute
150 Deokjin-dong, Yuseong-gu, Daejeon, 305-353, South Korea
**Corresponding author: hjryu@kaeri.re.kr*

1. Introduction

Oxide dispersion strengthened (ODS) steels are candidate materials for fuel claddings of Generation IV nuclear reactors as well as for the structural components of the fusion reactor due to its superior high temperature creep properties and good irradiation resistance. ODS steels are expected to be used under a long time high dose neutron radiation up to 200 dpa at an elevated temperature up to 973K, so the effects of a neutron radiation on mechanical properties and microstructural stabilities of ODS steels are primary concerns to be investigated. Since the presence of stable and fine oxide particles are critical to the good high temperature strength and creep properties of ODS steels, it shall be essential to understand and maintain the stability of the oxide particles under irradiation. This paper gives an overview of the recent progress in this field.

2. Microstructural features of ODS steels

The microstructure of ODS steels consists of a metal matrix with uniformly distributed oxide dispersoids, which are stable at a high temperature. The finely distributed oxides inhibit dislocation motions in the metal matrix and increase the resistance of the alloy to creep deformation. Another function of the dispersoid particles is to inhibit the recovery and recrystallization processes, which allow a very stable grain to be obtained. Studies showed two populations of oxides size-wise: very fine particles (few nms) vs. larger ones (few tens to hundreds of nms) in Y-Ti-O reinforced ODS steels. Fine nanoclusters have a very high O content while coarser particles appear to be $Y_2Ti_2O_7$ and TiO_2 or other oxides. The grain morphology of ODS steels is usually anisotropic and shows an elongated grain structure after hot extrusion or rolling process. ODS steels fabricated by the mechanical alloying (MA) method could contain a high density of defects as well as argon bubbles, which could serve as nucleation sites for microstructure change.

High dose irradiation may cause a dramatic evolution in a microstructure including the cavities in particles, "black dot" damages, dislocation loops, particle amorphization and a chemical composition change of oxide particles. Cavity, dislocation loops and black dots are frequently observed, while the amorphization has only been reported by a few researchers [1].

Akasaka et al. have carried out neutron irradiation experiments up to a dose of 15 dpa and found that there is no distinct microstructural change in any irradiated specimens of F94, F95(12Cr-2W-0.3Ti-0.24Y_2O_3, F94:MA in He gas, F95: MA in Ar gas) and M93(9Cr-2W-0.2Ti-0.35Y_2O_3) ODS steels [2]. It is also found that there was no change of the dislocation density and precipitates both within the grains or at grain boundaries in ferritic ODS steels, while there was a slight tendency for them to increase in the martensitic steels. The high resistance to radiation induced microstructural changes in the ODS steels was attributed to the oxide particles which can act as sinks for the point defects which are frequently produced under the neutron irradiation. Further investigation will be needed to make the evolution and the role of the interfaces under the neutron irradiation clearer.

3. Oxide particle stability under irradiation

According to Russell's equation for the time rate of change of the radius of a particle under radiation (Eq. (1)) [3, 4],

$$\frac{dr}{d\sigma} \cong -\frac{\xi}{N} + \frac{3D}{4\pi r K}\frac{C_t}{C_p} - \frac{Dr^2 n}{K}, \qquad (1)$$

where r is the particle radius; σ, the dose (dpa); K, the dose rate (dpa/s); ξ/N, the radius loss due to ballistic collisions due to radiation; D, the diffusion coefficient; C_t, the total solute concentration; C_p, the concentration of the solute in precipitates; ($C_t - C_p$, the remaining concentration in solution) and n is the density of precipitates (number per unit volume) in the matrix.

Precipitates can grow through diffusion of solute from the matrix (second term in Eq. (1)) or can shrink due to radiation-induced recoils (first term in Eq. (1)) or through release and diffusion of solute to other precipitates (third term in Eq. (1)). For a precipitate described by Eq. (1), large particles would shrink and small particles would grow, leading to a stable radius that depends on the dissolution constant (n), the displacement rate (K), and the diffusion coefficient (D).

It has been reported that the small-sized oxide particles tend to disappear under high doses (~200 dpa) of neutron and the large-sized precipitates were surrounded by a "halo" of small precipitates. However, this phenomenon is supposed to be a result of a long time in a high temperature reactor (400ºC) after irradiation [5]. According to the work by Allen et al. [4], as the total radiation dose increases, the average nanocluster size decreases and the number density of the nanocluster particles increases. Fu et al. investigated the mechanism of a high O solubility and a nucleation of nanoclusters in Fe and pointed out that a vacancy plays an indispensable part, which makes the formation energy of these nanoclusters lower and clusters stable [6]. Therefore, it could be possible that the particles dissolve into the matrix under irradiation and become smaller; at the same time, vacancies formed by the irradiation might lead to a precipitate of new stable nanoparticles and particle number density increases. For large oxide particles, amorphization and the particle size change were observed. Motta's work showed that a degradation of the particle interfaces (amorphization) starts rather early in the irradiation stage by Fe ions, but was not observed in the neutron irradiations (435 to 580ºC) of the same alloy and be attributed this difference to the difference in dose rate between the ion and the neutron irradiation[1].

Due to the different irradiation facilities and techniques, results about the stability of oxide particles are rather confusing. We have summarized recent data to get a 3-D figure (Figure.1) on temperature, dose and dose rate to clarify whether any trend exists with the oxide particle stability. It indicates that oxide particles become unstable as the irradiation temperature and the dose increases, while the dose rate seems to have smaller influence on the oxide stability. Motta et al. particularly studied the microstructure evolution of ODS steels (DY, M16, MA956, MA957) under a relatively low ion irradiation dose (12dpa) and at room temperature to elucidate a change in an oxide particle morphology (low left corner of the figure.1). Since tests at a higher temperature and dose (middle part of the figure.1) show that oxide particles remain stable, this abnormal data may be attributed to the slow recovery at a low temperature due to the low diffusion rate. This is consistent with Russell's theory prediction that particles would dissolve under radiation at lower temperatures because a sufficient diffusion cannot occur to re-precipitate.

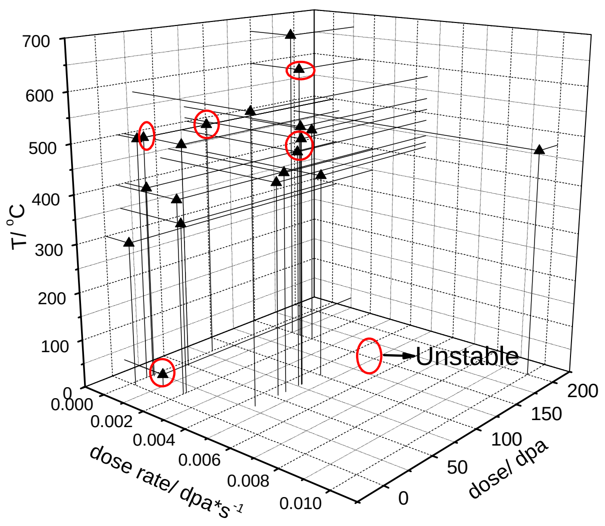

Figure.1. Literature survey of the stability of oxide particles vs. irradiation dose, dose rate and temperature

Among researchers focused on the stability of oxide particles, T.R. Allen, I. Monnet and T. Motta reported changes of oxide particles both at a low dose and a high dose, while Yamashita reported on a stable oxide particles irradiated in the experimental fast reactor JOYO at temperatures of 670-807K to 15.0 dpa maximum.

Monnet et al. pointed out that irradiation particle energy may play an important role as to whether oxide particles could dissolve into the matrix by electron irradiation [4]. It is possible that a minimum irradiation energy to displace O or Y/Ti ions from the oxide particle to dissolve it may exist.

4. Summary

The stability of oxide particles in ODS steels under irradiation at elevated temperature were analyzed by reviewing the various irradiation test results with electrons, heavy ions, neutrons or helium particles at different radiation doses, dose rates and with different energies. It indicates that the results on stability of oxide particles vary from each other, which may be due to the differences in the composition of the ODS steels concerned, the investigation methods and or the observation techniques. According to our survey, it's possible that at a high temperature & high dose irradiation, as well as at a low temperature & low dose irradiation oxide particles are prone to be unstable, while at a middle dose and middle temperature (300-400ºC) oxide particles stay stable, which is decided by the interaction between the dissolution by recoil and the particle growth by diffusion. Since the data about stability of an oxide particle in ODS steels are limited and variable, the effects of irradiation on an oxide particle need more intense study.

REFERENCES

[1] Arthur T. Motta, Robert C. Birtcher, Phase stability under irradiation of precipitates and solid solutions in model alloys and in ODS alloys relevant for GenIV, Report for phase III and final project report, 2007

[2] N. Akasaka, S. Yamashita and T. Yoshitake, Microstructural changes of neutron irradiated ODS ferritic and martensitic steels, Journal of Nuclear Materials, 329-333, 1053–1056, 2004

[3] K.C.Russell, Phase instability under cascade damage irradiation, Journal of Nuclear Materials, 206, 129-138, 1993

[4] T.R. Allen, J. Gan and J.I. Cole, Radiation response of a 9 chromium oxide dispersion strengthened steel to heavy ion irradiation, Journal of Nuclear Materials, 375, 26-37, 2008

[5] I. Monnet, P. Dubuisson and Y. Serruys, Microstructural investigation of the stability under irradiation of oxide dispersion strengthened ferritic steels, Journal of Nuclear Materials, 335, 311-321, 2004

[6] C.L. Fu, Maja Krcmar and G.S. Painter, Vacancy mechanism of high oxygen solubility and nucleation of stable oxygen-enriched clusters in Fe, Physical review letters, 225502, 1-4, 2007