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Microstructural changes of neutron irradiated ODS ferritic and martensitic steels

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Abstract

Oxide dispersion strengthened (ODS) ferritic and martensitic steels are promising materials for fuel cladding in fast reactors and for structural materials in fusion reactors. ODS steels for fuel cladding in fast reactors have been developed by Japan Nuclear Cycle Development Institute (JNC). The first generation ODS steels had a serious problem from anisotropy in their mechanical properties. Recently, this problem has been solved by controlling grain shape as equiaxed grains. Three types of modified ODS steels were irradiated in the experimental fast reactor JOYO for investigation of the irradiation-induced microstructural changes. The dispersed oxide particles were very stable when irradiated. In addition, dislocation structure development was minor. The high resistance to irradiation-induced microstructural changes in the ODS steels was attributed to oxide particles which acted as sinks for point defects.

1. Introduction

High swelling resistance and stable mechanical properties at elevated temperature under neutron irradiation conditions are very important issues not only for development of a long-life fuel assemblies in a fast reactors, but also for first wall materials in a fusion reactors. Irradiation performances of austenitic steels have been studied for nuclear plants. The results of these investigations, however, showed that the neutron dose limit in austenitic in a steels is around 120 dpa because of high swelling.

Ferritic steels, generally, have not only a fundamental characteristic of good swelling resistance up to very high neutron dose but also reduced long term radioactivity through adjustment of the chemical composition. On the other hand, it is necessary to improve the mechanical properties of ferritic steels for high temperatures under heavy neutron irradiation conditions, because the mechanical properties of ferritic steel are weaker than those of austenitic steel at high temperatures [1]. One promising way to improve the mechanical properties of ferritic steel is to get dispersion of fine and stable oxide particles in the matrix; known as oxide dispersion strengthened (ODS) ferritic steel.

Reduced activation ODS ferritic or martensitic steels for fuel cladding of fast reactors have been developed by Japan Nuclear Cycle Development Institute (JNC). The early generation ODS steels developed by JNC had a serious problem of anisotropy in the mechanical properties when used for cladding tubes, which was caused by elongated grains from drawing. Recently, controlling grain shape as equiaxed grains during manufacturing has solved this problem, and some manufacturing processes for making very usable ODS steels which have excellent high-temperature strength and cold workability were established [2–5].

Also, the neutron irradiation performance of the early generation ODS steels have been evaluated. In

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previous works, it has been shown that remarkable changes of mechanical properties and microstructures on the early generation ODS steels after neutron irradiation did not occur [6–8]. However, it is very important that the irradiation performance of these newly modified ODS steels is investigated, because there is a possibility that the irradiation behavior differs between early and modified ODS steels due to differences in structural morphologies.

The objective of this research is to investigate the irradiation-induced microstructural changes in the newly modified ODS steels, and assess the phase stability of the steels during neutron irradiation.

2. Experimental procedure

Three types of modified ODS steels without final cold working were prepared for investigation of microstructural changes during neutron irradiation. Their chemical composition is shown in Table 1. The first and second types of modified ODS steels had a recrystallized ferritic structures, called F94 and F95. The mechanical alloying processes of F94 and F95 were performed in helium gas and argon gas, respectively. The last type, M93, had an equiaxed crystal grain of martensite structure formed by α–γ phase transformation during thermal treatment. The mechanical alloying process for M93 was done in argon gas. The specimens prepared for neutron irradiation were short tubes with outer diameter and thickness the same as fuel pins for a fast reactor.

Neutron irradiation of ODS specimens was carried out in the experimental fast reactor JOYO. The environment during irradiation on the outer and inner specimen surfaces was liquid sodium. Irradiation temperature and neutron dose are shown in Table 2. After irradiation, TEM samples were prepared from irradiated specimens by mechanical grinding and electrical polishing for observation of microstructures. The observations were carried out using a transmission electron microscope as the JEOL JEM-4000FX.

3. Results and discussion

Typical microstructures of unirradiated and irradiated specimens of modified ODS steels are shown in Fig. 1. Dislocation density on F94 and F95 is very low because of the thermal treatment for recrystallization. The matrices have some coarse precipitates ranging in size from 30 nm to 500 nm. These precipitates are found to contain iron, chromium, titanium and tungsten from EDS analysis, and they seem to be some kind of carbides. The dislocation structure of M93 is fine with a high dislocation density. These precipitates formed not only in the grains, but also on the grain boundaries.

Table 1
Chemical composition of ODS ferritic and martensitic steels

Material	Chemical composition (wt%)															Notes
	C	Si	Mn	P	S	Ni	Cr	Ti	W	Y_2O_3	Ex. O	N	Ar	He	Fe	
F94	0.058	0.03	0.049	0.004	0.004	0.025	11.78	0.30	1.93	0.24	0.04	0.01	0.0003	0.0002	Bal.	MA in He gas
F95	0.058	0.03	0.049	0.004	0.004	0.025	11.78	0.30	1.93	0.24	0.04	0.01	0.0038	–	Bal.	MA in Ar gas
M93	0.12	0.02	0.036	0.003	0.004	0.022	8.99	0.20	1.94	0.35	0.06	0.01	0.0033	–	Bal.	MA in Ar gas

MA: mechanical alloying.

Table 2
Conditions of neutron irradiation

Temperature (K)	Neutron dose (dpa)
673	2.5
723	14
773	15
603	7.0

There is no distinct microstructural change in any irradiated specimens. The dislocation structure of M93 was stable following the irradiation. In F94 and F95, it is recognized that dislocation density increases slightly. It seems that the dispersed oxide particles lead to the high stability of the dislocation structures. In the case of M93, the oxide particles function mainly as obstacles to dislocation glide. In F94 and F95, the high interfacial oxide area retains a low point defect density in the matrix, because the interfaces act as strong point defect sinks.

It is also found that there is no change of precipitates both in grains and at grain boundaries in F94 and F95 during irradiation. In the case of M93, carbides show a slight tendency to form and to grow compared with the unirradiated condition. Also, another precipitate, possibly Laves phase, tends to forms near the interface between matrix and carbide following irradiation at 803 K.

Typical TEM images of dispersed fine oxide particles and cavities in unirradiated and irradiated specimens of modified ODS steels are shown in Fig. 2. In unirradiated specimens, many fine particles and some bubbles exist in the matrix. The fine particles with diameters around 10 nm are confirmed to be yttrium–titanium complex oxides by EDS analysis, and the dispersion of oxide particles in the matrix is uniform. Also, bubbles are uniformly distributed in the matrix. The bubble fraction is greatest in the order M93, F95 and F94.

After neutron irradiation, the differences in diameter and number density of bubbles between unirradiated and irradiated specimens of M93 are not clear. However, growth of bubbles is clearly recognized in F94 and F95, with bubble growth more conspicuous in F94, where the atmosphere during mechanical alloying was helium. It is possible that the bubble formation is, in principle, heavily dependent on impurity gas atoms which remain in the matrix from mechanical alloying, because the unirradiated specimens of all materials in this study have a lot of small bubbles. During neutron irradiation, bubbles growth may occur by the diffusion of the gas atoms, from interaction with vacancies and accumulation at point defect sinks which are dominated by the initial dislocation and oxide particle density. It may be that lower growth of bubbles in F95 (containing argon) compared with F94 (containing helium) is caused by slower diffusion of argon atoms, and the lowest growth

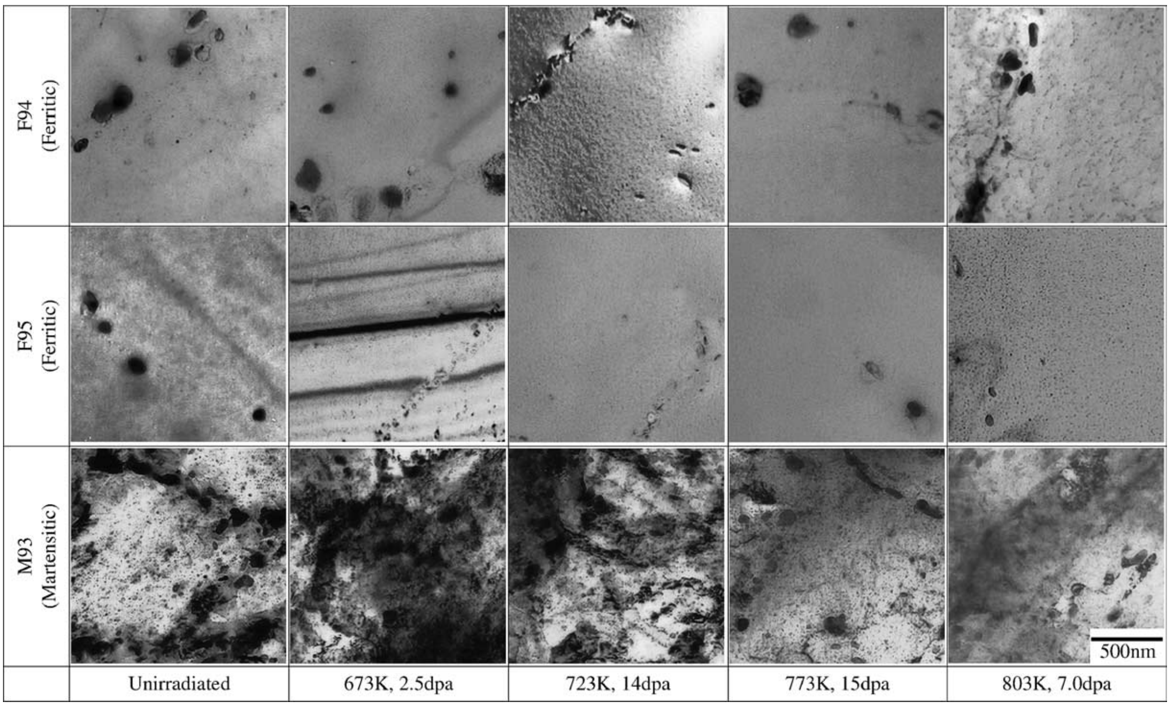

Fig. 1. Typical microstructures of each modified ODS steel.

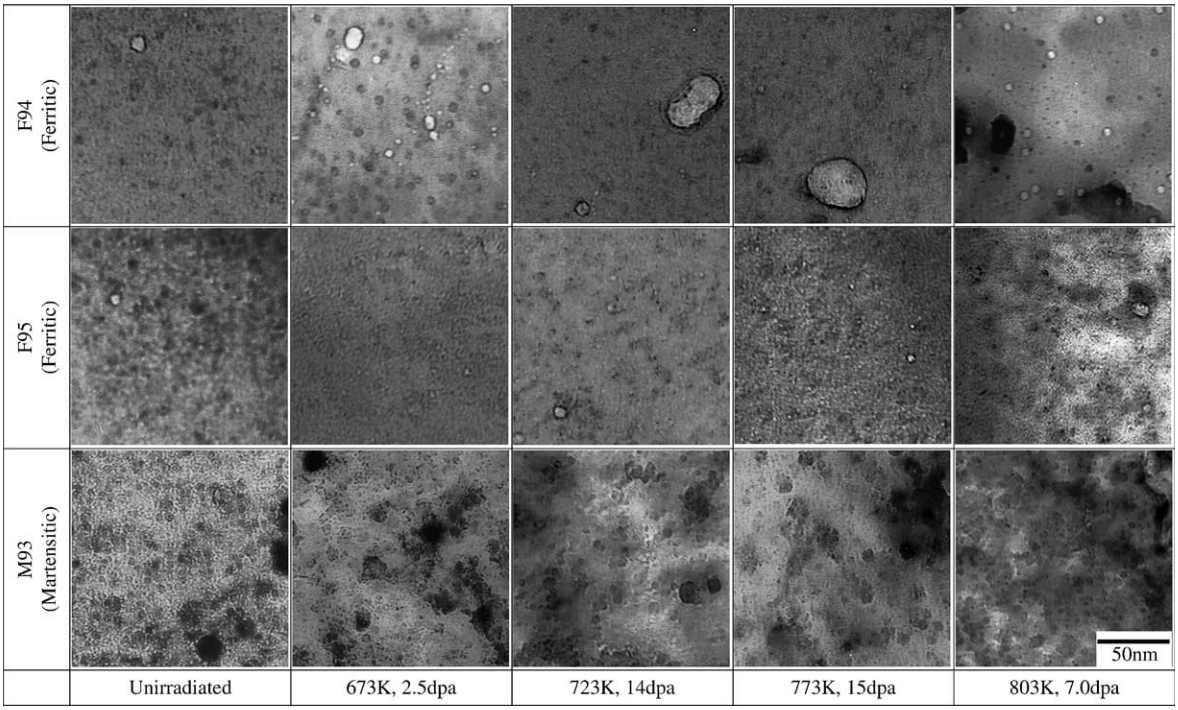

Fig. 2. Typical TEM images of dispersed fine oxide particles and bubbles in each modified ODS steel.

for bubbles for M93 may be due to the higher initial dislocation density.

The oxide particles dispersed in the matrix are stable during neutron irradiation. There is no evidence of growth and/or dissolution of the oxide particles. This means that ODS steels can be expected to retain their good mechanical properties and high resistance to irradiation induced microstructural changes. Details regarding the stability of the oxide particles are described elsewhere [9].

4. Conclusion

To determine phase stability of newly modified ODS steels which have equiaxed grains under neutron irradiation, microstructural observations were examined by TEM on three ODS steels irradiated in the experimental fast reactor JOYO. In general, all microstructures were stable under neutron irradiation. Therefore, it can be expected that these ODS steels perform well within the irradiation conditions in this experiment.

However, there is an effect from the gas, retainedfrom the mechanical alloying process, on cavity development. It is found that a heavier gas element is better than a lighter one in order to reduce irradiation-induced bubble coarsening.

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