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Shape memory characteristics of neutron irradiated Ti–Ni shape memory alloy couplers

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Abstract

Neutron irradiation effects on a Ti–Ni shape memory alloy (SMA) are not understood regarding the engineering behavior of tightening and connecting core parts. Irradiation response on shape memory capabilities for Ti–Ni SMA couplers was investigated by measuring the inner diameter of a SMA coupler. Ti–Ni SMA couplers were irradiated to a fast neutron fluence ($E > 1$ MeV) of 1.4×10^{25} m^{-2} at 323 and 503 K in the Japan Materials Testing Reactor of JAERI. The inner diameter versus temperature curves showed a large hysteresis for the SMA coupler irradiated at 323 K. At a neutron fluence above 10^{25} m^{-2}, the hysteresis after 323 K irradiation increased by 50% more than that after 503 K irradiation. The maximum amount of reversible stroke of the irradiated two-way Ti–Ni SMA coupler during heating and cooling was reduced to 70% that of an unirradiated one and the two-way Ti–Ni SMA coupler is more resistant to neutron irradiation than the one-way Ti–Ni SMA coupler. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

Ti–Ni shape memory alloys (SMA) have been considered to be useful for fission and fusion materials in which the shape memory effect, superelasticity and stress relaxation resistance [1] are applicable for the simple and quick replacement of coupled components, remote controlled operation and maintenance procedures and improving operational efficiencies in high radiation damage environments [2]. It was reported that neutron irradiation induced structural alterations in Ti–Ni SMA suppressed the martensitic transformation. On the other hand, the restoration phenomena from an irradiated state to an unirradiated state may also take place during irradiation and facilitate the martensitic transformation [3,4].

From a practical view point, it is demanded that the restoration phenomena in heavily irradiated Ti–Ni SMA coupler are understood. Those phenomena may give us useful information on the development of newly functional materials of high resistance to irradiation. It is important to clarify the effects of various irradiation conditions: temperature, fluence and fluence rate on the behavior of Ti–Ni SMA coupler. The purpose of this study is to investigate the relationship between the deformation behavior and the restoration phenomena for neutron irradiated Ti–Ni SMAs in order to understand the changes in the behavior of Ti–Ni SMA coupler after irradiation.

2. Experimental

Four kinds of ingots were prepared by using a high frequency vacuum melting furnace. The compositions were Ti–49.8, 50.0, 50.3 and 51.4 at.%Ni. The ingots were hot forged and cold rolled to produce sheets with a thickness of 1.0 mm and 10 mm for tensile and Charpy impact test specimens. Some of the forged ingots were cut and worked into SMA couplers. Tensile and impact specimens were homogenized at 1273 K for 3.6 ks and subsequently aged at 673 K for 3.6 ks followed by quenching into ice water. Coupler specimens were also homogenized at 1123 K for 1.8 ks and were expanded by

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inserting a punch for both one-way (by normal transformation of a parent phase to a martensitic phase) and two-way (by reversible transformation between a parent phase and a martensitic phase) couplers until 3.5% of the diameter of the coupler was attained. For two-way coupler specimens, constrained aging was also performed at 673 K for 0.17 Ms after expanding to memorize the parent phase and martensitic phase. Specimen sizes were $1 \times 10 \times 55$ mm^3 plate for tensile tests, $10 \times 10 \times 55$ mm^3 (normal size) and $5 \times 10 \times 55$ mm^3 (half size) bar for Charpy impact tests and an inner diameter of 6 mm and outer diameter of 10 mm for inner diameter measurement of SMA couplers, respectively. Transformation temperatures of specimens were determined by the differential scanning calorimetry (DSC) measurement. Cold worked to 20% reduction and laser-welded specimens were also prepared as reference materials for tensile and Charpy impact testing.

Neutron irradiation was carried out up to a fast neutron fluence ($E > 1$ MeV) of 1×10^{25} m^{-2} at temperatures of 323 and 520 K in the Japan Materials Testing Reactor (JMTR) of the Japan Atomic Energy Research Institute (JAERI). After irradiation, tensile tests were carried out for the irradiated and post-irradiation annealed specimens with a gauge length of 12 mm, at a strain rate of 7×10^{-4} s^{-1} at various temperatures between 153 and 333 K by using a remote controlled tensile machine. Tensile stresses were loaded to a strain of 5% and then, unloaded and heated above the A_f. temperature. Charpy impact tests were performed to measure the fracture energies of irradiated specimens in the temperature range between 153 and 373 K. The inner diameter of an SMA coupler was continuously measured using an R-type crip gauge at temperatures between 298 and 368 K to obtain transformation strains due to a martensitic transformation and the reverse transformation.

3. Results and discussion

3.1. Tensile properties of Ti–Ni alloys

Transformation temperatures for unirradiated alloys were determined by DSC measurements and shown in the table in Fig. 1. Transformation temperatures decreased with increasing Ni content. The M_s temperature of a cold worked alloy was lower than that of an aged alloy because of the presence of the stress field caused by dislocations as shown in the figure.

The effect of annealing at 473, 523 and 573 K after irradiation to a low fluence of 4.6×10^{22} m^{-2} was determined using the stress–strain curves for Ti–51.4 at.%Ni (aged), Ti–50.3 at.%Ni (laser-welded) and Ti–50.0 at.%Ni (cold worked) alloys at temperatures between 153 and 333 K. In order to examine the relationship between the deformation stress and the M_s temperature, the critical stresses (0.5% proof stresses) for annealed alloys to induce martensites are plotted as a function of deformation temperature in Fig. 1. The critical stresses of alloys annealed at 523 and 573 K can each be approximated by two straight lines, which intersect at a temperature close to the M_s temperature of post-annealed alloys. As shown in the figure, an anneal at 473 K still indicates a small restoring force, but the alloys become substantially restored after holding at temperatures above 523 K, as the annealing temperature is increased gradually. For this low fluence irradiation condition, the decrease in M_s temperature in a welded alloy was smaller than that in an aged one.

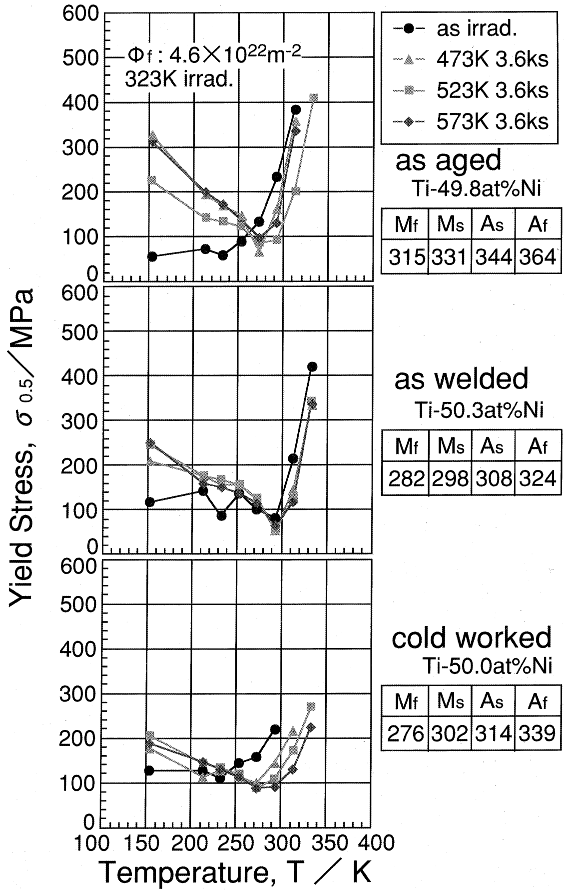

Fig. 1. Critical stresses (0.5% proof stresses) for irradiation to a neutron fluence of 4.6×10^{22} m^{-2} and post-annealing of alloys on the formation of stress-induced martensites as a function of test temperature.

3.2. Charpy impact properties of Ti–Ni alloys

Fracture energy versus temperature curves for Ti–Ni alloys irradiated to a fluence of 1×10^{25} m^{-2} at 330 K are shown in Fig. 2(a): fracture energies tend to decrease with increasing temperature regardless of prior treatment such as aging, cold-working and laser-welding, except for Ni rich Ti–51.4 at.%Ni alloy which contains a number of precipitates. Optical microscope observations of fracture surface on broken Charpy specimens showed fracture mode changes to markedly brittle behavior by the irradiation to a fluence of 1×10^{25} m^{-2}.

The dependences of fracture energies of Ti–Ni alloys on fluence and flux are shown in Fig. 2(b). Upon irradiation at 323 K to a fluence of 2×10^{22} m^{-2}, unusual impact properties were observed as described elsewhere [5], which can be explained as the sum of fracture energy curves of both parent phase and martensitic phase. At a fluence of 4×10^{22} m^{-2}, fracture energy decreased to a constant value of 4 J. When flux increased from 6.7×10^{16} to 1.6×10^{17} m^{-2} s^{-1}, a decrease in fracture energy was observed above 300 K. It was confirmed that the Ti–Ni alloy became brittle with increasing damage and the threshold value for the ductile to brittle transition corresponds to a fluence of 1×10^{23} m^{-2} and a flux of 1×10^{17} m^{-2} s^{-1}.

3.3. Changes in inner diameter of Ti–Ni shape memory alloy couplers

The inner diameter of the irradiated SMA couplers (types of one-way or two-way) was continuously measured for temperatures between 298 and 368 K during heating and cooling, since the gripping pressure of SMA coupler was inversely proportional to the inner diameter of the SMA coupler and also proportional to the thickness of the SMA coupler [6].

The inner diameter versus temperature curves shown in Fig. 3 indicates large hysteresis loop during heating and cooling of the SMA coupler irradiated at 323 K. At a neutron fluence of 10^{25} m^{-2}, the hysteresis of inner diameter of one-way or two-way couplers after 323 K irradiation is larger than that after 503 K irradiation as much as 50%, and therefore transformation temperatures could not be explicitly defined for one-way couplers from the inner diameter versus temperature curves. Decreases in the transformation temperatures and expansion of transition temperature region

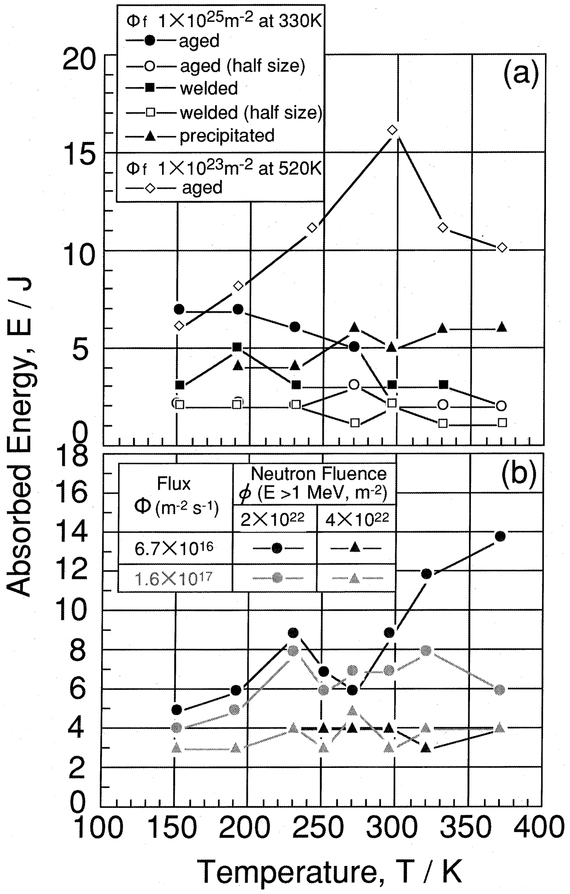

Fig. 2. Fluence and flux dependences of Charpy impact properties of Ti–Ni alloys irradiated to a neutron fluence of 10^{25} m^{-2}.

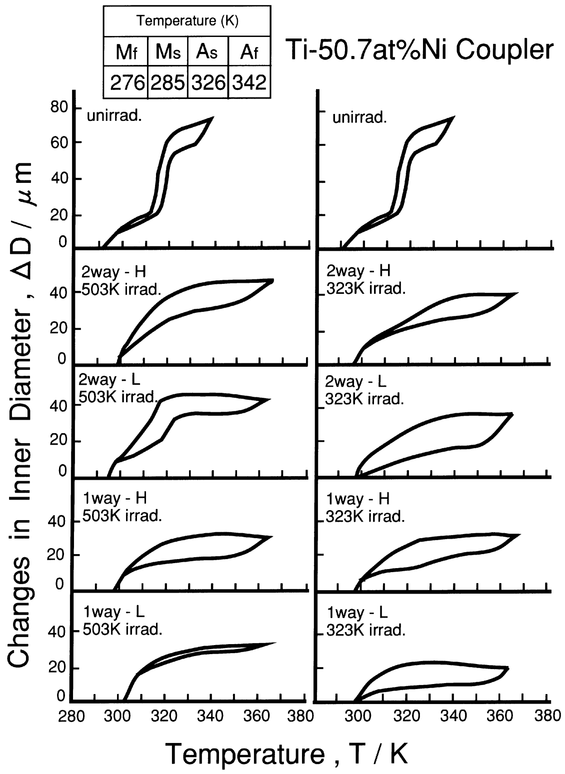

Fig. 3. Inner diameter versus temperature curves before and after irradiation for Ti–Ni shape memory alloy couplers.

take place in Ti–Ni alloy response after irradiation and can bring about wide hysteresis during heating and cooling in the inner diameter versus temperature curves. This indicates that the effect of neutron fluence (10^{25} m^{-2}) on shape memory capabilities of one-way Ti–Ni SMA coupler is greater than for the effect of irradiation temperature (503 K) under these irradiation conditions.

In the case of the two-way Ti–Ni SMA couplers, the maximum amount of reversible stroke for the irradiated two-way coupler during heating and cooling was reduced to 70% of unirradiated case which indicates that two-way couplers can have somewhat better resisitance to irradiation than one-way couplers. It can be seen that the presence of an internal stress field in two-way couplers is closely associated with the reduction of irradiation induced residual strains, which were measured by means of X-ray diffractometry [1].

The order–disorder transformation under irradiation can be described by the Bragg–Williams' degree of the order of "S" [7]. The derivative of the degree of long range order S with respect to time t in a disordering process can be expressed as a product of disordering efficiency and displacement rate times S. The derivative dS/dt in an ordering process can also be obtained as the product of $(1 - S)^2$ times K, which is a function of the vacancy concentration, the ordering jump frequency of vacancies and the temperature. The degree of long range order under irradiation is calculated from the boundary conditions of $dS/dt = 0$ [8].

Fig. 4 shows the change in the degree of long range order in equilibrium for a Ti–Ni alloy as a function of displacement rate (ϕ/dpa s^{-1}) at temperatures of 370 and 520 K for various displacements (Φ/dpa) [9]. As shown in the figure, the degree of order decreases at displacement rates of 10^{-6} to 10^{-8} dpa s^{-1}, when the irradiation is performed at 370 K. When the irradiation temperature is higher than 500 K, restoration phenomena occur in the alloy and the degree of order within the same range of displacement rates, which correspond to a fluence of $10^{17} m^{-2}$ s^{-1} at 520 K, is found to be above 0.5. Experimental results in Figs. 1 and 2 can be explained by the calculated results shown in Fig. 4.

The restoration phenomena for which ordering predominates over disordering occurs at temperatures around 520 K. The degree of order in equilibrium depends on the displacement level. At a low displacement of 10^{-2} dpa, the degree of order is expected to increase to 0.9, and the value is lowered with increasing displacement and is considered to be 0.4 at a displacement of 10^{-1} dpa which is almost equal to a fluence of 10^{23} m^{-2}. Above 1 dpa dose level, the degree of order in equilibrium is estimated to be zero and disordering becomes predominant.

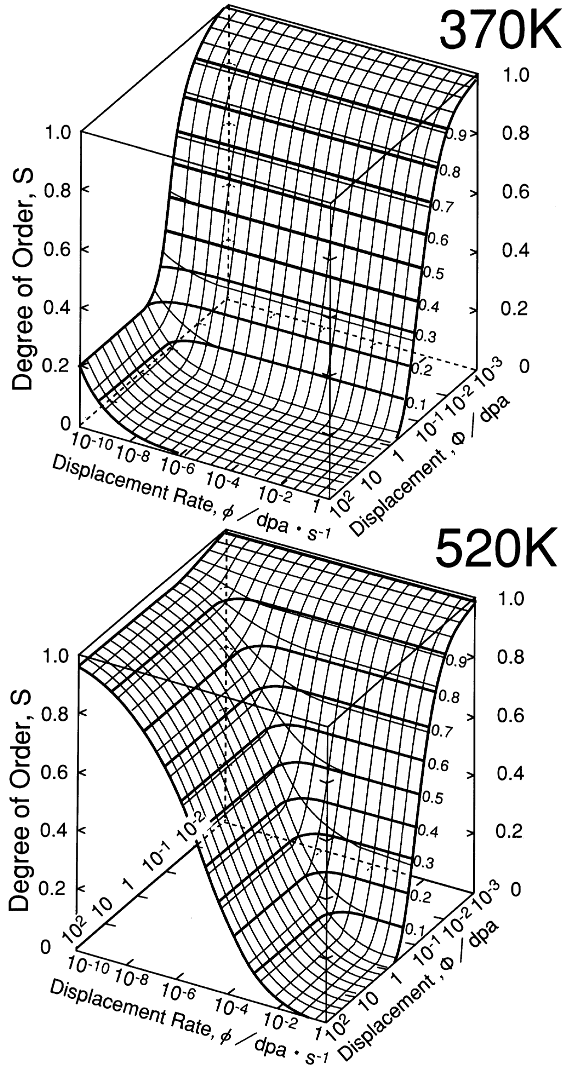

Fig. 4. Degree of long range order in equilibrium for Ti–Ni alloys as a function of displacement rate at temperatures of 370 K and 520 K for various displacements.

4. Conclusions

Shape memory effects have been investigated for Ti–Ni alloys after the irradiation at various temperatures to a maximum fast neutron fluence of 1×10^{25} m^{-2} and subsequent annealing at 473, 523 and 573 K. Main results are:

1. Regardless of prior treatment such as aging, welding and cold-working, Ti–Ni alloys return to a substantially restored state by post-irradiation annealing at temperatures above 523 K.
2. From fracture energy versus temperature curves after 330 K irradiation to a fluence of 1×10^{25} m^{-2}, threshold values of ductile to brittle transition correspond

to a fluence of 1×10^{23} m^{-2} and a flux of 1×10^{17} m^{-2} s^{-1}.

3. From the inner diameter versus temperature curves for SMA couplers, decrease in M_s temperature and expansion of the transformation temperature region can bring about a wide hysteresis. A reversible stroke of two-way couplers was reduced to 70% that for the unirradiated case at most, suggesting that two-way couplers having internal stress fields may be more resisitant to irradiation than one-way couplers.

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