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Tensile properties and hardness of two types of 11Cr-ferritic/martensitic steel after aging up to 45,000 h

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ABSTRACT

The relationship among tensile strength, Vickers hardness and dislocation density for two types of 11Cr-ferritic/martensitic steel (PNC-FMS) was investigated after aging at temperatures between 400 and 800 °C up to 45,000 h and after neutron irradiation. A correlation between tensile strength and Vickers hardness was expressed empirically. The linear relationship for PNC-FMS wrapper material was observed between yield stress and the square of dislocation density at RT and aging temperature according to Bailey–Hirsch relation. Therefore, it was clarified that the correlation among dislocation density, tensile strength and Vickers hardness to aging temperature was in good agreement. On the other hand, the relationship between tensile strength ratio when materials were tested at aging temperature and Larson–Miller parameter was also in excellent agreement with aging data between 400 and 700 °C. It was suggested that this correlation could use quantitatively for separately evaluating irradiation effects from neutron irradiation data containing both irradiation and aging effects.

1. Introduction

The ferritic/martensitic (F/M) steels are expected to be used not only for the long life core material of fast reactors (FRs) but also for the blanket materials of fusion reactors because of their superior swelling resistance. Two types of 11Cr-F/M steel (PNC-FMS) have been developed in the Japan Atomic Energy Agency as materials for wrapper tubes and cladding tubes for use in the Japanese Sodium-cooled FR (JSFR) [1–5]. These wrapper and cladding materials have the same range of elemental compositions, but their final heat treatments are different. Wrapper material was normalized and tempered at relatively low temperature to obtain the required high tensile strength. On the other hand, cladding material was normalized and tempered at higher temperature to achieve the required high creep strength. Toloczko and Garner [6–7] examined the swelling behavior of two different heat treated HT9 tubes after irradiation in FFTF/MOTA. These studies showed that small differences in strains associated with both phase-related changes in lattice parameter and void swelling were observed in comparing the two heats. Uehira et al. [1] examined tensile properties of PNC-FMS cladding and wrapper materials after accelerating aging at 410–750 °C up to 10,000 h They reported that tensile properties of two heats at the same aging temperatures were quite different. These previous studies [1, 6–7] have provided fundamental knowledge that irradiation and thermal aging behavior of F/M steels in different heat-treatments were different and therefore identical evaluations were difficult. However, it is not easy to acquire systematic irradiation data for materials with the same composition and heat-treatment conditions. Therefore, the focus of this study was to evaluate the relationship among tensile strength, Vickers hardness and dislocation density at room temperature (RT) with PNC-FMS wrapper material aged at 400–700 °C up to 15,000 h and cladding materials aged at 400–800 °C up to 45,000 h in order to investigate effective utilization of materials having different heat-treatments.

Previously Uehira et al. [1], calculated the strength of thermally aged specimens at various aging temperatures using the Larson-Miller parameter (LMP) of accelerated aging tests although it was known that the change of microstructures in the ferritic heat resisting steels, such as PNC-FMS, depended on aging temperatures. Therefore, in this study, the tensile tests at aging temperatures between 400 and 700 °C corresponding to normal and transient conditions in JSFR were also performed in order to separate the aging effects from irradiation effects after neutron irradiation.

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Table 1
Chemical compositions (wt%) and heat treatment conditions of PNC-FMS wrapper and cladding (representative lot).

Material (lot)	C	Si	Mn	P	S	Ni	Cr	Mo	W	V	Nb	N	Fe
Range (max/min)	0.15/0.09	<0.1	0.8/0.4	<0.03	<0.03	0.6/0.2	12.0/10.0	0.7/0.3	2.3/1.7	0.25/0.15	0.08/0.02	0.07/0.03	bal.
Wrapper (lot 22WFK)	0.13	0.05	0.80	<0.001	<0.001	0.52	11.02	0.40	1.86	0.22	0.06	0.06	bal.
Cladding (lot 61FS)	0.10	0.07	0.54	0.002	0.002	0.32	11.05	0.45	1.89	0.21	0.055	0.04	bal.

Wrapper: normalized at 1050 °C for 40 min and tempered at 710 °C for 40 min
Cladding: normalized at 1100 °C for 10 min and tempered at 780 °C for 60 min

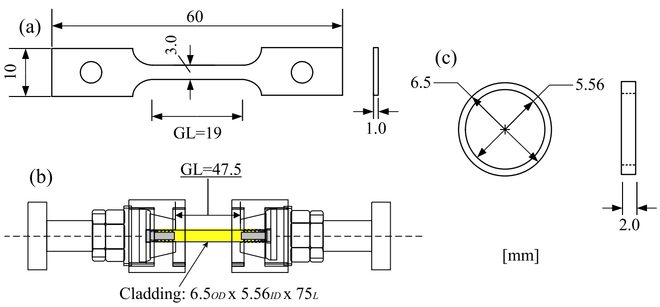

Fig. 1. Dimensions of tensile specimens. (a) Wrapper material specimen, and cladding specimens viewed in the (b) longitudinal direction and (c) transverse direction.

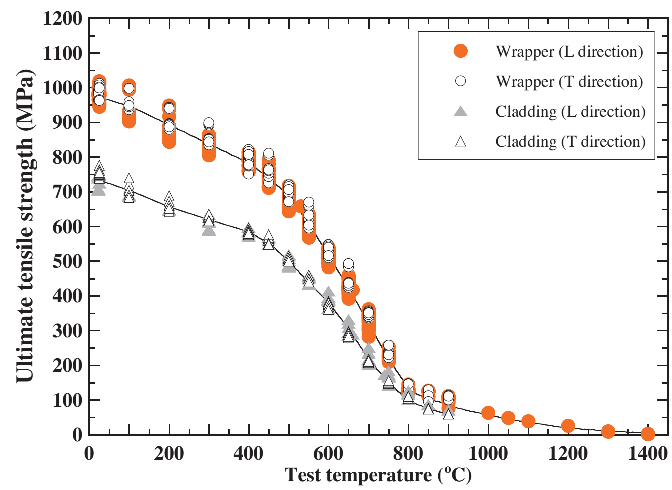

Fig. 2. Ultimate tensile strength of the two directional specimens for PNC-FMS wrapper and cladding materials.

2. Experimental

Materials used in this study were two types of PNC-FMS, one was developed as a wrapper tube material and the other as a cladding material. The PNC-FMS wrapper material was in the form of plates which were produced by the thermos-mechanical process equivalent to the wrapper tube fabrication process. The microstructures of wrapper and cladding are fully martensitic structures and martensitic structures with δ-ferrite less than 5%, respectively. The elemental compositions and heat treatments of the materials are listed in Table 1.

Thermal aging tests of PNC-FMS wrapper and cladding were carried out at temperatures between 400 and 700 °C up to 30,000 h and between 400 and 800 °C up to 45,000 h, respectively. PNC-FMS wrapper material was irradiated in the experimental FR Joyo using the core material irradiation rig (CMIR) at temperatures between 400 and 660 °C to fast neutron fluences ranging from 0.5 to 7.1×10^{26} n/m^2 (E>0.1 MeV), which was equivalent to 2.5–35.5 dpa.

Micro-Vickers hardness tests were conducted at the center of thickness direction with a load of 500 gf for 10 s before and after thermal aging for both materials.

The tensile specimens were machined from plates of the PNC-FMS wrapper material and cladding tubes in both longitudinal (L) and transverse (T) directions to the rolling direction; dimensions are as shown in Fig. 1. Tensile tests were carried out at RT, and at aging and irradiation temperatures in the air using a screw-driven tensile test machine at an initial strain rate of 5.0×10^{-5} s^{-1}, which was changed to 1.25×10^{-3} s^{-1} after yielding, according to JIS G0567, except for the O-ring tensile specimen as shown in Fig. 1(c). Yield strength (YS) was determined as 0.2% offset proof stress. The O-ring tensile tests according to T direction of cladding tubes were performed at constant cross-head speed of 0.1 mm/min to get only ultimate tensile strength (UTS) because it is very difficult to determine the gauge length accurately for the O-ring specimen due to non-uniform deformation. Therefore, as a result, O-ring tensile data except for UTS were unusable.

The two types of directional specimens were tested at each test temperature to examine effect of rolling on tensile properties, but the effect was negligible as shown in Fig. 2. Therefore, both longitudinal and transverse directional specimens were regarded to have similarity in this paper.

Dislocation density of wrapper material before and after aging was measured in the center of thickness direction by X-ray diffraction (XRD), using a modified Williamson-Hall plot as well as a modified Warren–Averbach plot [8–12]. For XRD analysis, the diffraction profiles of the (110), (200), (211) and (220) reflections were measured using a conventional diffractometer with Co Kα_1 and Kα_2 radiation operating at 40 kV and 135 mA at scan speeds of 0.1, 0.4, 0.5 and 0.04 (° min^{-1}), respectively. $\bar{C}_{h00}$ is the average dislocation construction factor corresponding to the (h00) diffraction and is determined by the elastic constants; $\bar{C}_{h00} = 0.285$ for pure iron was used in the present analysis [10]. The details of XRD profile analysis have been reported elsewhere [11,12].

3. Results and discussion

3.1. Vickers hardness, tensile properties and dislocation density at room temperature

Figs. 3(a) and (b) show the average Vickers hardness values and Vickers hardness ratio before and after aging as a function of thermal aging temperature, respectively. There was no significant change before and after aging below 550 °C. Both wrapper and cladding materials behaved in a similar manner. On the other hand, above 600 °C, the hardness values obviously decreased with increasing aging time; the decreasing ratio of wrapper material was much higher than that of cladding material for the same aging time. It was well known that the static recovery behavior of tempered martensitic structures can be evaluated by the hardness measurement [13]. In this regard, it was suggested that the Vickers values of both materials were not the same even if the aging time had been completed and that dislocation density in the wrapper

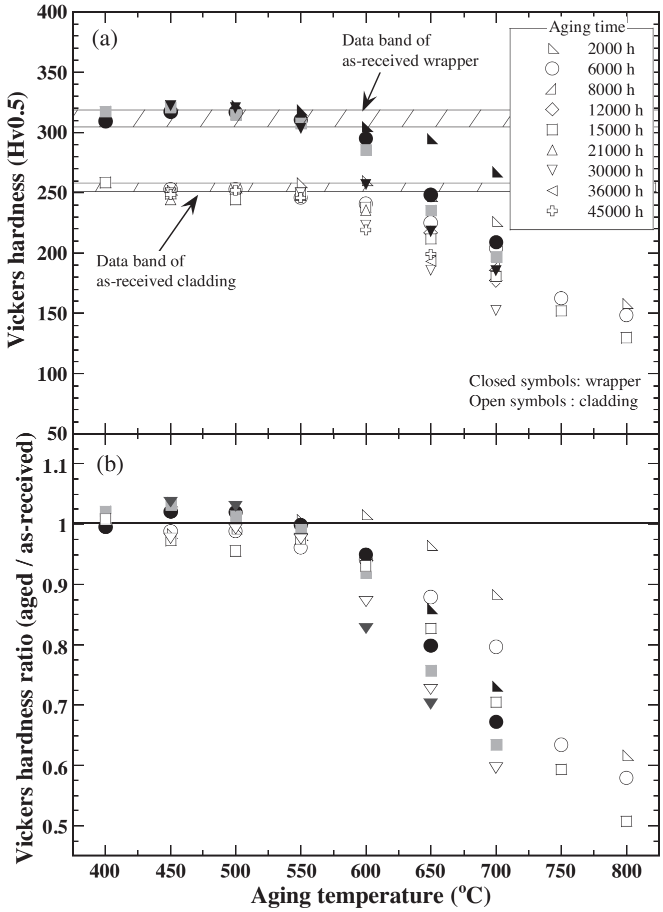

Fig. 3. Two properties as a function of aging temperature: (a) Vickers hardness and (b) Vickers hardness ratio.

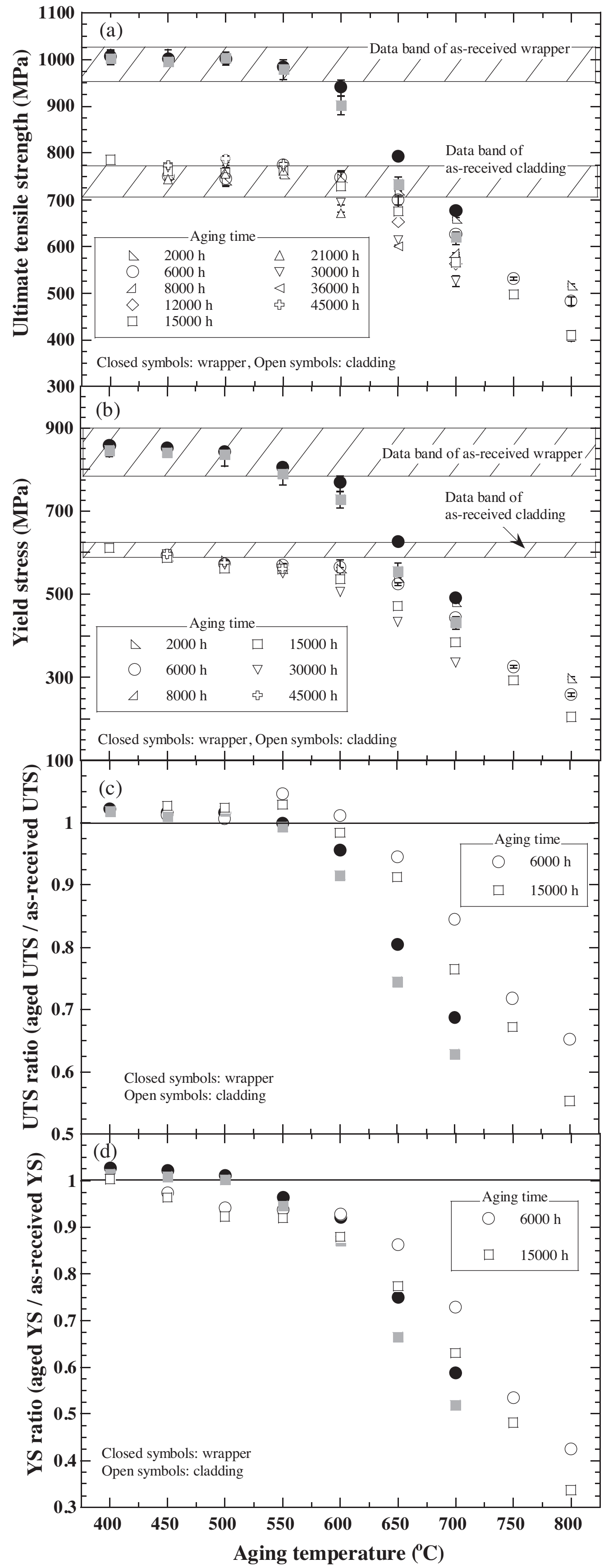

Fig. 4. Four properties as a function of aging temperature: (a) UTS, (b) UTS ratio, (c) YS and (d) YS ratio tested at RT.

material was higher than that of cladding material although recovery of martensitic structures in cladding changed relatively little.

The UTS and YS as a function of aging temperature are shown in Figs. 4(a) and (b), respectively. The decrease in UTS and YS due to aging are shown as the ratio of tensile strength before and after aging in Figs. 4(c) and (d), respectively. The trends of UTS and YS ratios were not treated as equivalent in the comparison of wrapper and cladding materials. This tendency was similar to that of Vickers hardness results. The reason would be that the recovery of martensitic structures in the wrapper material occurred but not for the cladding material due to a high density of dislocations according to different heat-treatment temperatures.

Busby et al [14] proposed a method to estimate the yield stress in irradiated steels from measured hardness, which was based on the concept of representative strain introduced by Tabor [15]. The present study extended beyond YS to UTS. Fig. 5 shows the relationship between Vickers hardness, YS and UTS. The number of YS and UTS data in Fig. 5 differed because O-ring tensile data except for UTS were unusable. The linear relationship was expressed empirically as follows $\sigma_y = 3.39\,\mathrm{Hv}–246.02$, $\sigma_u = 3.04\ \mathrm{Hv} + 11.59$, where σ is in MPa and Hv in kg/mm^2. The data corresponded to linear relationship although behaviors of cladding and wrapper were different. This result suggested that not only YS but also UTS could be evaluated by Vickers hardness beyond certain limits and that the irradiation effects on tensile strength at RT also could be estimated from Vickers hardness tests on PNC-FMS of different heat-treatment condition.

Fig. 6(a) shows the peak deconvolution of (110) reflection in the as-received wrapper material as a representative result. Each peak

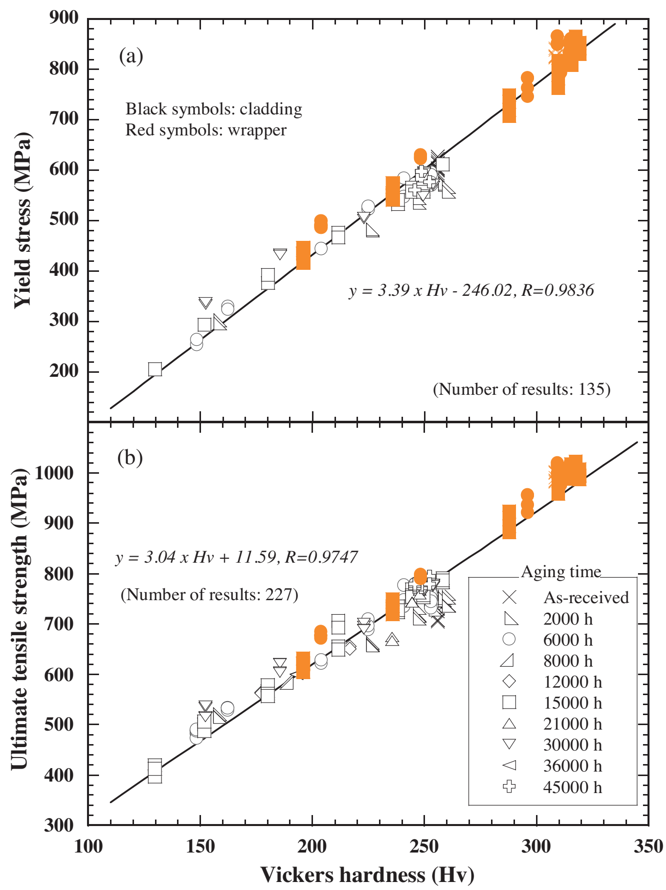

Fig. 5. The relationships (a) between YS and Vickers hardness and (b) between UTS and Vickers hardness.

was separated into $K\alpha_1$ and $K\alpha_2$ using the Lorentz function and the full width at half maximum (FWHM) was estimated from the fitting procedure. Fig. 6(b) shows the modified Warren–Averbach plot. As a result, $\rho = 3.87 \times 10^{14}$ m^{-2} was determined from the linear relationship as shown in Fig. 6(c). Fig. 6(d) shows the dislocation density in wrapper material as a function of aging temperature. As shown there, the decrease in dislocation density was observed above 550 °C and the recovery of martensitic structures was quantitatively evaluated by the change of dislocation density, although the dislocation density data varied widely. This decreasing tendency in the dislocation density was similar to that of the yield strength for wrapper material as shown in Figs. 4(b) and (d). In general, dislocation density (ρ) and flow stress (τ) can be expressed using Bailey–Hirsch relation [16]

$$\tau = \tau_0 + \alpha \mu b \rho^{-1/2} \tag{1}$$

where, α and τ_0 and b are constants and μ is the shear modulus. Therefore, Fig. 7 shows YS as a function of square root of dislocation density for the as-received and aged wrapper material. The linear relationship was expressed empirically as follows: $\sigma_y = 2.32 + 4.14 \times 10^{-5} \sqrt{\rho}$, where σ_y is in MPa and ρ in m^{-2}. This correlation was approximately linear with slight scatter among the data. When b of pure iron and μ of PNC-FMS at RT were 0.248 nm and 83.89 GPa, α, which included Taylor factor, became 1.15. Supposing that the Taylor factor of bcc is 3.06 [17], true-α can estimate 0.376. This linear relationship was seemed to be valid because it was well known that the values of α was typically 0.2–0.4 at RT without dependence on materials [18]. Thus, it was clarified that the correlation among dislocation density (recovery of martensitic structure), tensile strength and Vickers hardness was in good agreement and that Micro-Vickers hardness tests were useful as a quantitative measure of mechanical and microstructural properties in materials of different heat-treatments after aging and after irradiation.

3.2. Tensile properties at aging and irradiation temperatures

Figs. 8(a)-(d) show YS, UTS, uniform elongation (UE) and total elongation (TE) before and after aging and irradiation as a function of test temperature, where in addition to the results of irradiated PNC-FMS wrapper, the other reported data of PNC-FMS cladding (lot 61FS) irradiated in Joyo[2–3] are given for comparison. Irradiation hardening of wrapper material occurred at low irradiation dose, but a significant decrease in elongation did not occur below 400 °C. For cladding material, there was no significant degradation in tensile strength before and after aging and/or irradiation between 400 and 550 °C as shown in Figs. 8(a) and (b). On the other hand, above 550 °C, the decrease in tensile strength for wrapper material occurred even after aging. Furthermore, after aging above 650 °C, there was significant decrease in tensile strength during aging and/or irradiation. Meanwhile, the change of elongation occurred even although there was no significant change of tensile strength after irradiation as shown in Figs. 8(c) and (d). These results suggested that effects of irradiation on tensile properties were reflected by elongation rather than strength. Then, we used the Bailey-Hirsch relation, Eq. (1), in order to evaluate the main factor influencing tensile strength at high temperatures. In this relation, α ,τ_0 and b are constants without dependence on test temperature, but the shear modulus changes according to test temperature. Fig. 9 shows YS results obtained at 550, 600, 650 and 700 °C as a function of (shear modules at each temperature [19]) times (square root of dislocation density at RT). The correlation in this figure was roughly linear with some scatter of the data. The linear relationship assumed that a parameter of temperature dependence for Bailey-Hirsch relation in this situation had significant influence on μ rather than τ_0, which contains thermal stress components such as Peierls stress, although it might be suggested that the same τ_0 correspond exactly to each test temperature. In the case of PNC-FMS, Laves and $M_{23}C_6$ carbides, which are precipitates distributed on the grain boundaries and in the grain interiors, might be considered as another microstructural factor affecting the tensile strength. The co-authors obtained the result showing that the number density of Laves phase was highest at 550–600 °C and Laves phase size was largest at 700 °C in the PNC-FMS wrapper materials aged at 400–700 °C. These indicated that the main factor governing the tensile strength at high temperatures was the stability of dislocation structures, which meant stability of lath martensitic structures. Therefore, it was suggested that tensile strength at high temperatures would be estimated by measuring Vickers hardness at RT. The authors are presently carrying out aging tests up to 90,000 h and will continuously accumulate aging and/or irradiation data to evaluate these hypothesis.

Figs. 10(a) and (b) show YS and UTS ratios (aged and irradiated strength / nominal as-received strength) as a function of LMP, respectively. The nominal curves in these figure are the regression lines from all experimentally aged wrapper and cladding data between 400 and 700 °C as well as data of the previous study (excluding accelerated aging data over 700 °C) [1]. The trend of the YS ratio was different from that of the UTS ratio in both materials and the decreasing in the YS ratio was larger than that in the UTS ratio. Furthermore, the decreasing trend of the ratio for cladding and wrapper materials was also different. These results were different from those of the previous study [1]. It was suggested that the difference between this and the previous study would be attributed to both the number of data accumulated and no data for

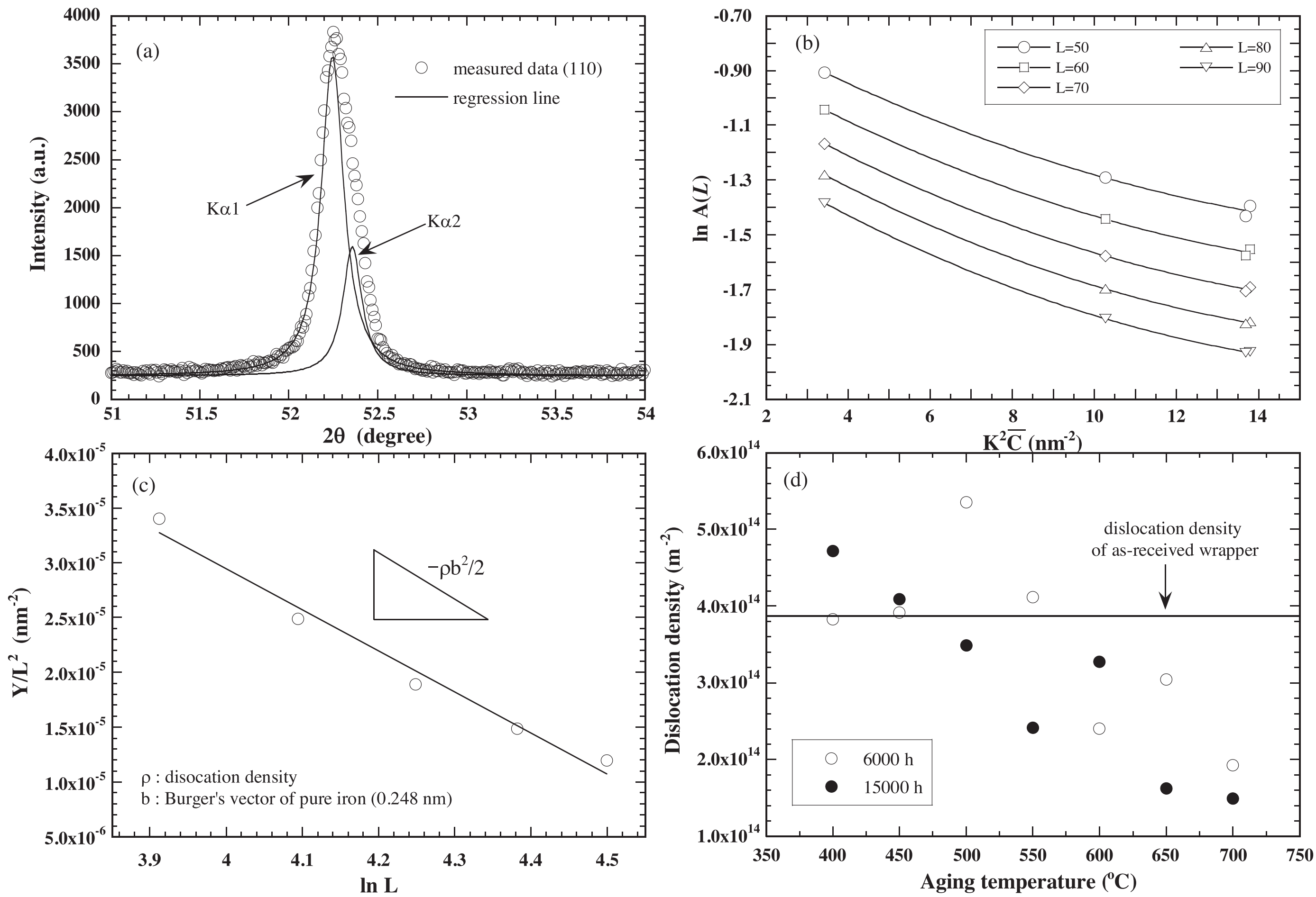

Fig. 6. (a) The peak deconvolution of (110) reflection, (b) the modified Warren-Averbach plot and (c) the relationship between *ln*L (L: Fourier Length) and Y/L2 for as-received PNC-FMS wrapper. (d) The dislocation density of PNC-FMS wrappers before and after aging.

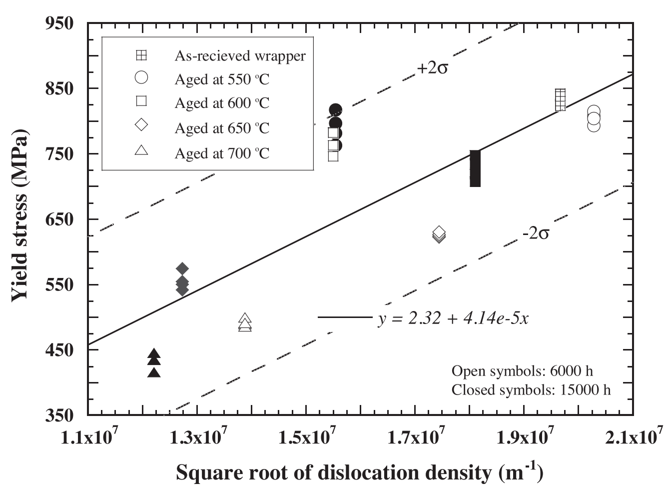

Fig. 7. Correlation for wrapper material between square root of dislocation density and YS tested at RT.

accelerated aging tests. This relationship between tensile strength ratio and LMP was in excellent agreement with aging data and it was suggested that this correlation could use quantitatively for separately evaluating irradiation effects from neutron irradiation data containing both irradiation and aging effects.

4. Conclusions

Tensile tests, Vickers hardness tests and dislocation density measurements of two types of aged PNC-FMS were conducted at RT to investigate the influence of different heat-treatments on the effective utilization of the materials. After aging and irradiation, tensile tests were carried out at various aging and irradiation temperatures to separate the aging effects from irradiation effects. The results are summarized as follows:

(1) Even though the PNC-FMS materials had different heat-treatments, there was single empirical correlation between tensile strength and Vickers hardness as follows: $\sigma_y = 3.39$ Hv–246.02, $\sigma_u = 3.04$ Hv + 11.59, where σ is in MPa and Hv in kg/mm^2.

(2) The linear relationship for PNC-FMS wrapper materials was observed between YS and the square of the dislocation density at RT and aging temperature according to the Bailey-Hirsch relation. This result indicated that the main factor governing the tensile strength was the stability of dislocation structures.

(3) It was clarified that the correlation among dislocation density, tensile strength and Vickers hardness to aging temperature was in good agreement. Vickers hardness tests were useful as a quantitative measure of mechanical and microstructural properties for the different heat-treatment materials.

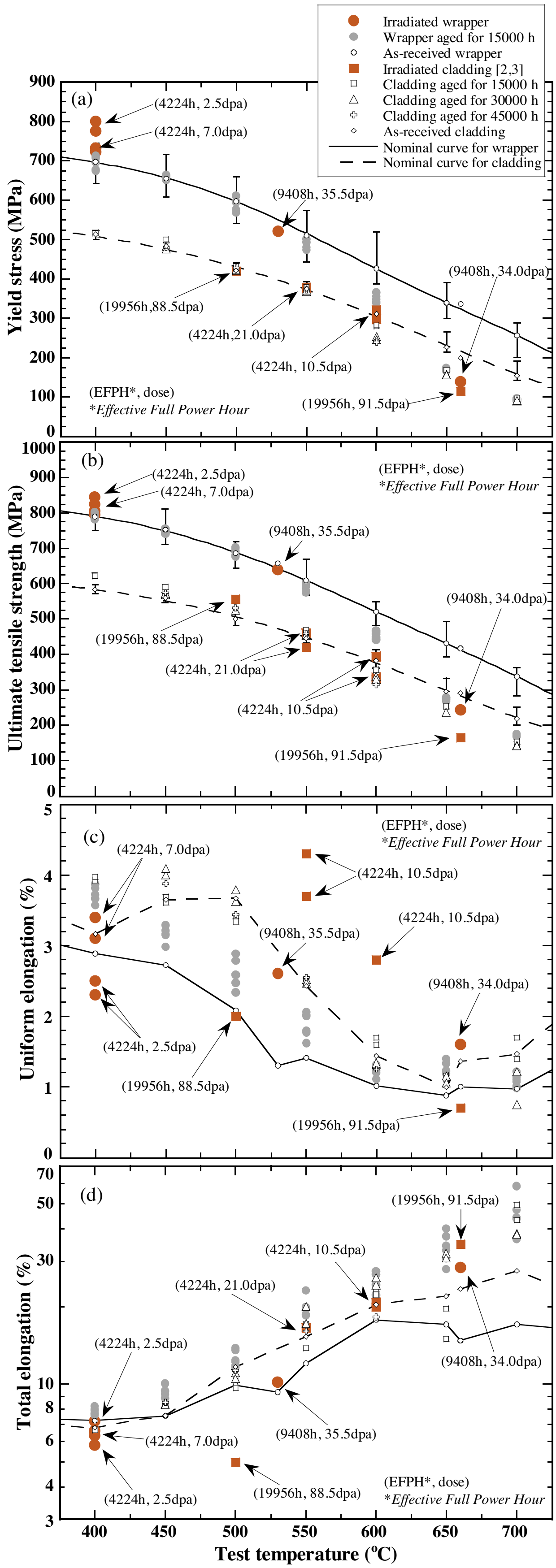

Fig. 8. Four properties as a function of test temperature: (a) YS, (b) UTS, (c) UE and (d) TE.

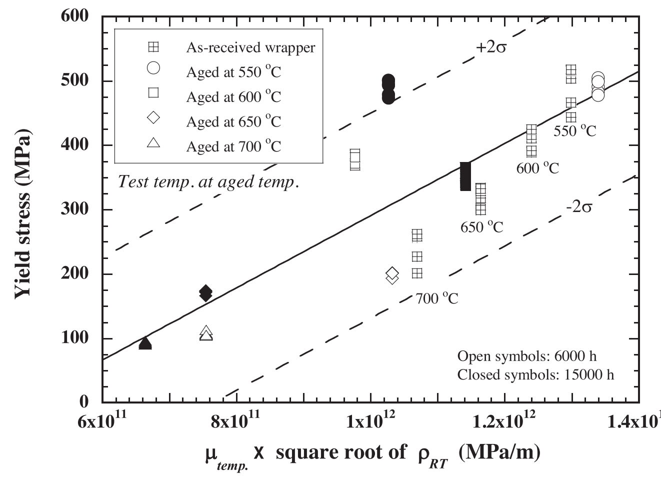

Fig. 9. YS results for wrapper material at various aging temperatures as a function of (shear modulus) times (square root of dislocation density).

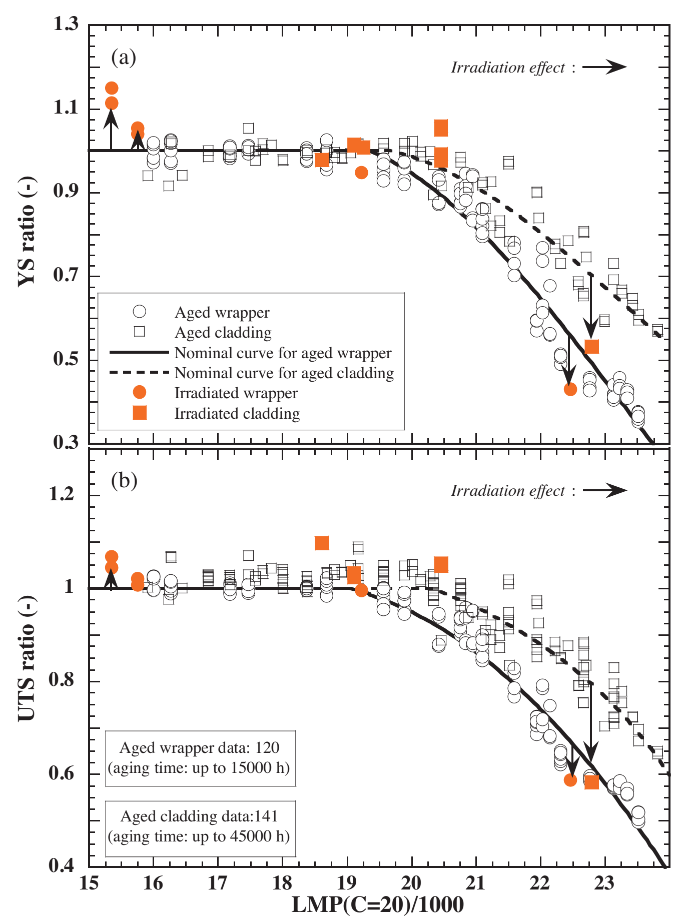

Fig. 10. Relationships for PNC-FMS wrapper and cladding materials: (a) YS ratio vs. LMP(C = 20), and (b) UTS ratio and LMP (C = 20).

(4) The relationship between tensile strength ratio and LMP was in excellent agreement with aging data between 400 and 700 °C corresponding to normal and transient conditions in JSFR. The irradiation effects could be quantitatively separated from neutron irradiation data by this correlation.

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