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Effect of thermal aging on the microstructure and mechanical properties of 7–11 CrW steels

Y. de Carlan [a,*], A. Alamo [a], M.H. Mathon [b], G. Geoffroy [c], A. Castaing [a]

[a] CEREM/SRMA, CEA Saclay, 91191 Gif sur Yvette Cedex, France

[b] Laboratoire Léon Brillouin, CEA-CNRS, Saclay, 91191 Gif/Yvette Cedex, France

[c] Laboratoire des Solides Irradiés, Ecole Polytechnique, 91128 Palaiseau, France

Abstract

The aim of this paper is to present the evolution of the microstructure, tensile and impact properties of Fe–7.5/11CrWVTa reduced activation martensitic (RAM) steels after thermal aging performed at relevant temperatures for fusion applications (250–550°C). The materials investigated are experimental alloys with different contents of Cr (7.5–11%), Ta (0–0.1%), W (0.8–3%) and interstitial elements, such as carbon (0.09–0.17%) and nitrogen (0.004–0.045%). Thermal aging was performed for up to 13 500 h on steels with different metallurgical conditions: normalized and tempered (N&T) and normalized and tempered + 10% cold worked (N&T-CW). The characterization of the microstructure was performed by transmission electron microscopy (TEM) and by small angle neutron scattering (SANS). The evolution of tensile properties depends essentially on the initial metallurgical conditions, whereas the modification of impact properties is mainly defined by the chemical composition that governs the occurrence of alpha prime and Laves phase precipitation.

1. Introduction

Reduced activation martensitic (RAM) steels are candidates for internal structures of fusion reactors. The qualification of this type of material implies knowledge of their behavior under irradiation in the range of temperatures envisioned for fusion reactors (250–550°C). It also implies a comprehension of the aging mechanisms and the possibility to distinguish between the thermal contribution and the radiation damage.

The stability of the microstructure and mechanical properties is directly related to the chemical composition and the initial metallurgical condition (tempering, cold working, etc.). Thus, the objective of this work is to perform a systematic study to analyze the influence of some elements (W, Cr, Ta, N, C) and the metallurgical condition on the behavior of different RAM steels after thermal aging. The results are compared to those obtained on conventional FeCrMo(VNb) steels, which are the base for RAM steel development.

The experimental RAM steels studied in this work are a series of six alloys developed and supplied by AEA-Culham and two melts – F82H and JLF-1 from, respectively, JAERI and Tokyo University. The physical metallurgy including the effects of chemical composition, heat treatments and cold working on prior austenite grain size, phase transformation characteristics and mechanical properties on as-received (AR) materials have already been studied and reported [1,2]. This work presents the microstructural and mechanical evolution of these RAM steels after thermal aging performed in the range 250–550°C to simulate in-service conditions. Mechanical characterization on aged samples was carried out by tensile and Charpy-V tests. The microstructure was also characterized by transmission electron microscopy (TEM) and by small angle neutron scattering (SANS) to analyze the relationships between chemical composition, mechanical properties and microstructure.

* Corresponding author. Tel.: +33-1 69 08 61 75; fax: +33-1 69 08 71 30.

E-mail address: yann.decarlan@cea.fr (Y. de Carlan).

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2. Materials and experimental procedures

The six steels produced by AEA-Culham (named LA etc.) with different contents of Cr, W, Ta, C and N were forged as plates of 26 mm thickness and finally hot and cold rolled to a thickness of 3.5 mm. The last fabrication step consisted of a normalization treatment for 40 min at 1030°C, a tempering treatment for 1 h at 750–795°C and finally 10% cold rolling. So, the initial metallurgical condition of these RAM steels is a normalized + tempered + cold-worked martensitic structure (N&T-CW).

The F82H and JLF-1 steels have been supplied as plates 7.5 and 15 mm thick in the normalized and tempered conditions (N&T), that is, normalization at 1040°C (F82H) and 1050°C (JLF-1) and tempering at 750°C (F82H) and 780°C (JLF-1) for 1 h. The chemical composition, prior austenite grain size and metallurgical conditions of the steels are given in Table 1. Thermal aging was performed under vacuum (10^{-2} Torr) for up to 5000 h for LA12LC and LA12TaLC steels, 10 000 h for LA12TaLN, LA12Ta, LA13Ta and LA4Ta steels and 13 500 h for F82H and JLF-1 steels.

Cylindrical tensile specimens with a 2-mm diameter and 12-mm gauge length were machined parallel to the rolling direction. The tensile tests were performed in air at a strain rate of 3×10^{-4} s^{-1} and the test temperatures ranged from 20°C to 650°C.

Impact specimens were machined along the rolling direction (LT orientation). They were Charpy-V-notch subsize specimens (27 mm long, 4 mm wide and 3 mm thick) for LA12LC and LA12TaLC and quasi-standard Charpy-V-notch samples of 55 mm long, 10 mm wide and 3.5 mm thick for the other steels. Charpy-V tests were conducted over the temperature range from −200°C to +400°C, in order to produce full transition curves for each material. The ductile–brittle transition temperature (DBTT), corresponding to 50% ductile and 50% cleavage fracture mode, was deduced from the half-value of the Upper Shelf Energy (USE) or from force–time curves. The accuracy of DBTT values is about ±10°C. Energy values were normalized to the initial cross-section area (3×3 mm^2 for the subsize specimens and 8×3.5 mm^2 for the quasi-standard specimens).

3. Microstructural characterization

The microstructure of RAM steels consists of laths of tempered martensite within prior austenite grains. Only in F82H steel, some regions partially recrystallized are observed (see Fig. 1) in the AR condition as well as in the aged specimens. For each alloy, different kinds of precipitates were identified in carbon extraction replicas (LA12LC, LA12TaLC and LA12TaLN steels were not characterized by TEM). For all the steels, the main precipitation consists of $M_{23}C_6$ precipitates of variable size (10–200 nm) with a typical composition of metallic elements of 60Cr–30Fe–5V–5W (in at.%). These carbides are located along grain and lath/subgrain boundaries. Depending on the alloys (in F82H steel only $M_{23}C_6$ were identified), other carbonitrides were observed in the AR condition:

- M_2X: Cr-rich in LA4Ta steel;
- MX or M_6X: Ta-rich in LA4Ta, LA12Ta, LA13Ta and JLF-1 steels;
- M_4X_3: V-rich particules in LA4Ta, LA12Ta, LA13Ta and JLF-1 steels.

After thermal aging, no significant evolution of the precipitation was observed by TEM except in Cr-rich LA4Ta steel where M_2X particles disappeared, whatever the aging temperature was, and in W-rich LA13Ta steel, where Fe_2W type Laves phase was observed after aging at 550°C for 10,000 h. This last phase appears as a thin film that engulfs carbides along grain and lath/subgrain boundaries (see Fig. 2). The chemical composition of this Laves phase film obtained by XEDS was 49Fe–24Cr–2V–25W (in at.%). The occurrence of the Laves phase was already observed in that steel after thermal aging but at higher temperature (500 h – 675°C) [3].

Table 1
Chemical composition (wt%) and metallurgical features of RAM steels[a]

Alloy	C	Si	Mn	Cr	V	W	N	Ta	G.S. (μm)	Met. cond.
LA12TaLN	0.165	0.02	0.84	9.04	0.24	0.75	0.0048	0.10	20	N&T + CW
LA12Ta	0.155	0.03	0.88	9.86	0.28	0.84	0.0430	0.10	20	N&T + CW
LA13Ta	0.179	0.04	0.79	8.39	0.24	2.79	0.0480	0.09	25	N&T + CW
LA4Ta	0.142	0.03	0.78	11.08	0.23	0.72	0.0410	0.07	7	N&T + CW
LA12LC	0.089	0.03	1.13	8.92	0.30	0.73	0.035	0.01	40	N&T + CW
LA12TaLC	0.090	0.03	1.13	8.80	0.30	0.73	0.019	0.10	25	N&T + CW
F82H	0.087	0.10	0.21	7.46	0.15	1.96	0.0066	0.023	100	N&T
JLF-1	0.106	0.05	0.52	8.70	0.18	1.91	0.028	0.08	25	N&T

[a] G.S. – Prior austenite grain size (μm); Met. cond. – Initial metallurgical condition.

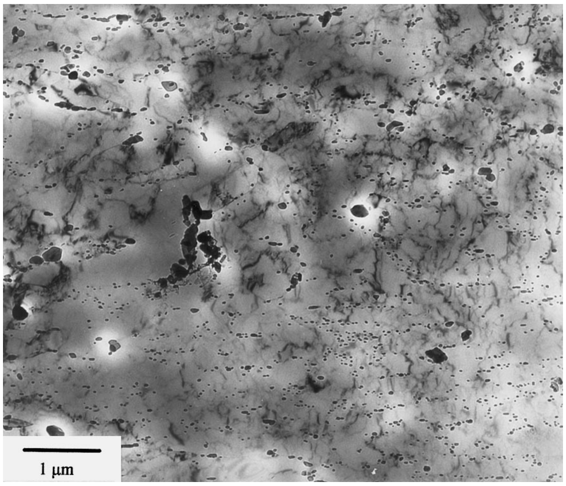

Fig. 1. TEM micrograph of a partially recrystallized zone in the AR F82H steel.

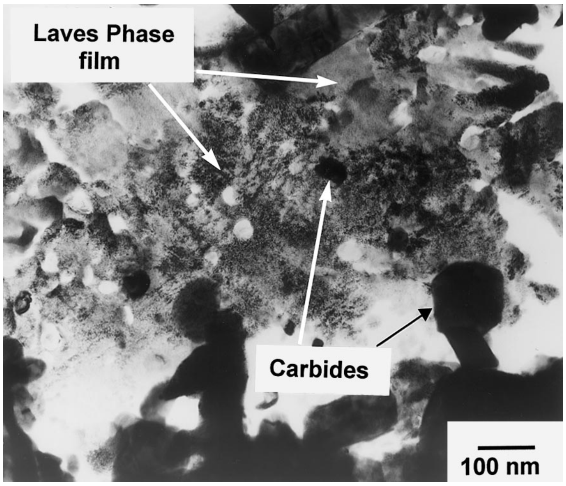

Fig. 2. TEM micrograph of Laves phase that engulfs carbides – carbon extraction replica of 9Cr 3WVTa steel aged for 10 000 h at 550°C.

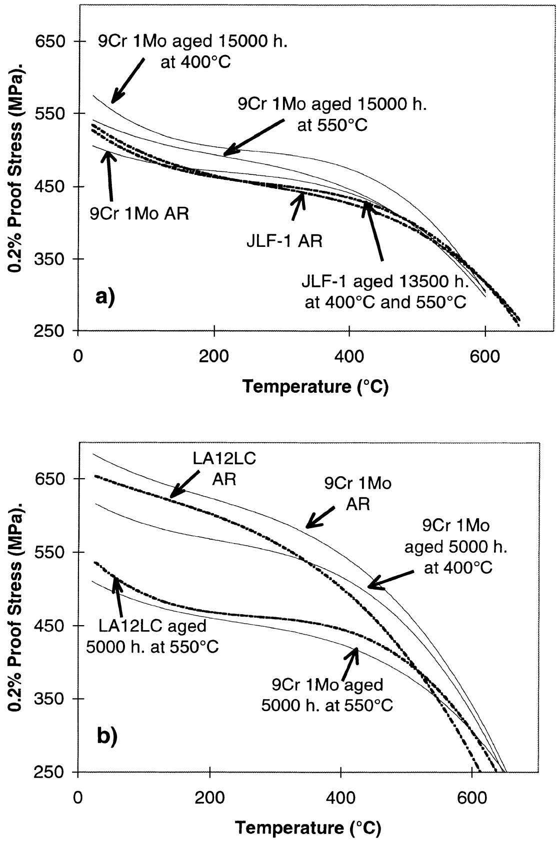

Fig. 3. Evolution of the 0.2% proof stress with thermal aging of steels having different initial metallurgical conditions. (a) N&T initial condition; (b) N&T-CW initial condition.

4. Mechanical behavior

Before aging, strength values of all the materials were similar to those of the conventional 9Cr–1Mo steels obtained in the same metallurgical condition [2]. As shown in Fig. 3, for LA12LC steel and JLF-1 steel, the evolution of tensile properties after thermal aging depends on metallurgical conditions. Cold-worked steels exhibit a decrease in the proof stress because of the recovery of the structure and no significant change of ultimate tensile strength (UTS) and ductility is observed. This fact indicates the enhancement of strain hardening capacity of N&T-CW steels, which reached low initial values. N&T steels display very stable strength values, and just a slight decrease of the reduction in area (RA) is detected specially after aging at 550°C.

Table 2 gives the impact properties of all the steels studied. Impact properties are not very sensitive to the initial metallurgical conditions, but depend on chemical composition and on aging temperature. The main modifications are observed after aging at 400°C in the case of the highest Cr-content (LA4Ta alloy) and at 550°C for high W-concentration (LA13Ta steel).

An empirical correlation between RA values and USE values was found (see Fig. 4). In fact, RA values measured from tensile tests at 20°C and 450°C show, for a given Charpy-V-notch specimen geometry, a linear dependence with experimental USE values. All metallurgical conditions are included: AR, N&T, N&T-CW and aged at different temperatures and times. This correlation allows a good estimation of the USE value to be obtained from the measurement of the RA after a tensile test at 20°C. The same correlation (with the same coefficients as shown in Fig. 4) is observed for conventional FeCrMoVNb steels [4].

Table 2
USE and DBTT of RAM steels before and after aging[a]

Aging temperature	USE (J/cm^2)						DBTT (°C)					
	AR	250°C	350°C	400°C	450°C	550°C	AR	250°C	350°C	400°C	450°C	550°C
LA12TaLN[b]	170		150	155	155	160	−65		−75	−55	−70	−70
LA12Ta[b]	190		180	145	155	180	−60		−70	−70	−65	−70
LA13Ta[b]	145		135	135	135	110	−70		−50	−45	−65	−20
LA4Ta[b]	155		130	130	140	140	−65		−65	−45	−65	−65
LA12LC[c]	100	105	105	110	100	110	−75	−85	−75	−75	−75	−90
LA12TaLC[c]	100		100	110	105	110	−70		−75	−80	−70	−85
F82H[d]	195	200	205	190	200	175	−75	−65	−50	−55	−55	−30
JLF-1[d]	190	190	180	190	185	160	−75	−65	−60	−70	−60	−45

[a] AR: as-Received.
[b] 10 000 h aged, quasi-standard Charpy-V-notch specimens.
[c] 5000 h aged, Charpy-V-notch subsize specimens.
[d] 13 500 h aged, quasi-standard Charpy-V-notch specimens.

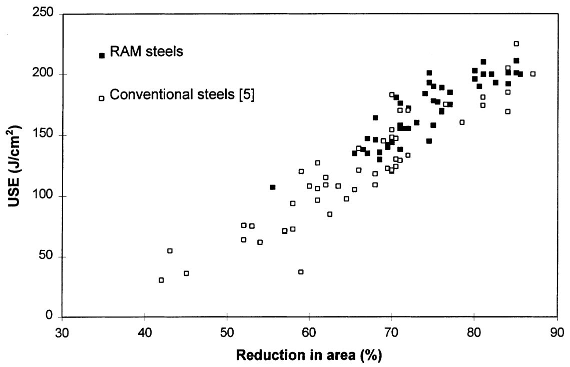

Fig. 4. Correlation between the RA to failure (determined from tensile tests at 20°C) and USE (determined from Charpy-V tests – quasi-standard Charpy-V-notch specimens).

After aging at 400°C, in the case of steel with the highest Cr-content (11%), a degradation of impact properties is observed. TEM observations in LA4Ta (11Cr–WVTa) steel show that thermal aging between 400°C and 550°C seems to induce the dissolution of M_2X particles (which were initially located between $M_{23}C_6$ carbides). This evolution does not seem to be able to explain the embrittlement observed in that steel. On the other hand, SANS experiments carried out on LA4Ta aged at 400°C show the occurrence of alpha prime particles all over the matrix, which could explain this behavior [5]. As in conventional FeCrMoVNb martensitic steels, the increase of Cr-content induces a detrimental effect on impact properties after thermal aging in the temperature range from 400°C to 450°C [4,6].

Concerning the aging at higher temperature (550°C), the behavior depends on the W-content in the alloy. No degradation of impact properties is observed for materials with 0.7% W aged at 550°C. For a W-content above or equal to 2% (F82H and JLF-1), the USE decreases and the DBTT increases. For the steel with 3% W (LA13Ta), a clear embrittlement is observed, i.e., a decrease of 20% of the USE and an increase of 50°C of the DBTT associated with indications of intergranular fracture mode. This behavior, due to the precipitation of Laves phase as an intergranular brittle film, is also observed in conventional 9/12Cr–MoW martensitic steels [4,7]. According to Tamura et al. [8] and Abe et al. [9], the temperature where the kinetics for Laves phase precipitation are fastest occurs at about 650°C in 9Cr–4W and 7.5Cr–2W steel and is slightly higher compared to that of Laves phase formation in 9/12Cr1Mo steels (T91 and HT9) [8].

In F82H and JLF-1 steels (7.5/9 Cr–2WVTa), a clear embrittlement (increase of the DBTT of about 30–45°C) was detected after aging at 550°C. On the other hand, after 13 500 h, at this aging temperature, the precipitation of Laves phase was neither observed by TEM nor by SANS [5], even though it was observed in these steels after thermal aging at 650°C [8], fatigue tests at 600°C [10] and neutron irradiation at 460°C, 600°C and 750°C [11–13]. Before the precipitation of Laves phase at grain/lath boundaries, one can assume the presence of some segregation of W at interfaces (as observed by Lapeña et al. [14] in F82H aged at 500°C), which is the early stage of Laves phase precipitation responsible for the embrittlement observed after thermal aging at 550°C.

The aging treatments performed on RAM steels in this study does not allow to precisely evaluate the influence of the interstitial content (C, N) because of the important contribution of Cr and W contents which mask the effects of the interstitial elements.

5. Conclusion

Experimental Fe–7.5/11CrWVTa RAM steels were characterized by mechanical tests and by TEM to obtain data on the evolution of properties after thermal aging.

The evolution of mechanical behavior after aging depends on the chemical composition and metallurgical condition (N&T or N&T-CW).

Steels with Cr $\leqslant$ 9% and W $\leqslant$ 1% exhibit no degradation of mechanical behavior after aging in the range from 250°C to 550°C. Cold-worked materials enhance their strain hardening capacity during aging.

Two different embrittlement mechanisms were detected which are related to the chemistry of alloys:

- At 400°C for materials with Cr $\geqslant$ 10% due to the occurrence of α' particles. This precipitation is strongly accelerated by irradiation [5].
- At 550°C for materials with W $\geqslant$ 2%. This behavior is explained by the segregation of W at grain boundaries and/or precipitation of Laves phase.

The Ta-content in the materials allows the control of the grain size in the austenitic field, but does not seem to influence directly the mechanical properties after thermal aging.

Finally, an empirical correlation is proposed that allows an estimate of the USE of a RAM steel from the reduction in area measurements after a tensile test.

The results obtained in this work show a similar behavior between FeCrWVTa RAM steels and conventional FeCrMoVNb martensitic steels before and after aging.

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