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Effect of Heat Treatments on Precipitate Microstructure and Mechanical Properties of CuCrZr Alloy

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Abstract

A number of specimens of CuCrZr alloy was prime aged and then overaged at 600°C for 1, 2 and 4 hours and for 4 hours at 700 and 850°C. After different heat treatments, both the precipitate microstructure and mechanical properties were characterized. Mechanical properties were determined at 50 and 300°C. Some selected specimens in the prime aged as well as overaged conditions were irradiated in the BR-2 reactor at Mol at 60 and 300°C to a displacement dose level of ~0.3 dpa. Irradiated specimens were mechanically tested at 60 and 300°C. The post-deformation microstructure of the irradiated specimens was examined in the transmission electron microscope. Results of the present investigations are presented in section 3. A brief summary of the main results and conclusions is given in section 4.

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1 Introduction

Currently, the precipitation hardened CuCrZr alloys are being evaluated for their use in the first wall and divertor components of ITER. In service, both these components will be exposed to an intense flux of fusion (14 MeV) neutrons and at the same time will experience thermo-mechanical stresses. Experimental investigations have demonstrated, however, that the CuCrZr alloy when irradiated with neutrons in the prime aged condition at temperatures below about 200^{o}C become harder but looses its ability to work harden and suffers from loss of ductility and plastic instability [1, 2]. Experimental results indicated that this may be due to the fact that the precipitates in this alloy in the prime aged condition were too small in size and thus too weak obstacles to dislocation motion during deformation. It was, therefore, decided to coarsen the precipitate size by annealing after prime ageing heat treatment so that the larger and thereby stronger precipitate may prevent the initiation of plastic flow localization by resisting dislocation motion.

Another reason for investigating the effect of heat treatment was the uncertainty in the temperature at which the copper alloys may be joined to 316 stainless steel while manufacturing the first wall panels. For this reason it was decided to investigate the effect of annealing at different temperatures that may be relevant for the manufacturing of the components. In the following, we describe these two sets of investigations on the effects of heat treatments on the precipitate microstructure (section 3.1). Main results of the post-deformation (after irradiation) microstructure are described in section 3.2. The mechanical properties of the CuCrZr alloy are described in sections 3.2 and 3.4. A brief summary and conclusions are presented in section 4.

2 Materials and Experimental Procedure

The CuCrZr alloy (Cu-0.73%Cr-0.14%Zr) supplied by Outokumpu Oyj (Finland) was solution annealed at 960^{o}C for 3 h, water quenched and then prime aged (PA) at 460^{o}C for 3 h. After prime ageing the specimens were given further heat treatments to modify the precipitate microstructure. In the first series, the prime aged specimens were annealed in vacuum at 600^{o}C for 1, 2 and 4 h. In the second series a number of prime aged specimens were annealed at 700 and 850^{o}C for 4 h. In both series of experiments, the specimens were water quenched after final annealing at various temperatures.

After each heat treatment, specimens were examined in a transmission electron microscope (TEM) in order to determine the effect of various heat treatments on the size and density of precipitates.

A number of tensile and fracture toughness specimens of the CuCrZr alloy in the prime aged condition and the specimens further heat treated at 600^{o}C for 1 and 4 hours were irradiated with fission neutrons in the BR-2 reactor at Mol (Belgium) at 60 and 300^{o}C to a displacement dose level of ~0.3 dpa (displacement per atom). The damage rate during irradiation was ~6 x 10^{-8} dpa s^{-1}.

The prime aged and various heat treated specimens were tensile tested at 50 and 300^{o}C. All tests were carried out in vacuum (<10^{-4} torr) at a strain rate of 1.2 x 10^{-3} s^{-1}. The single edge notched bend SEN(B) fracture toughness specimens of dimensions 3x4x27 mm were used in the fracture resistance testing. The initial notch and the 20% side grooves were machined by applying the electric wire discharge machining. The applied prefatigued crack length to specimen width ratio (a/W) was about 0.5. Fracture resistance curves were determined using the displacement controlled three point bend test method with a constant displacement rate of 1.5x10^{-2} mm/min. Fracture resistance testing at elevated temperatures were carried out in a silicon oil bath. Load, displacement and crack length measured using the DC-PD method were recorded during the testing and the fracture resistance curves were determined following the ASTM E1737-96 standard procedure.

The prime aged as well as specimens heat treated at 600^{o}C for 1 and 4 h and subsequently irradiated at 60 and 300^{o}C were also tensile tested in the post-irradiation condition. All tests were carried out in vacuum (<10^{-4} torr) at a strain rate of 1.2 x 10^{-3} s^{-1}. The specimens irradiated at 60^{o}C were tensile tested at 22 and 60^{o}C whereas those irradiated at 300^{o}C were tensile tested at 300^{o}C. Fracture toughness tests were carried out at 20 and 300^{o}C.

Characterization of the microstructures of CuCrZr specimens after different heat treatments was performed using a JEOL-2000FX transmission electron microscope. 3 mm diameter discs were punched out from the heat treated samples and were mechanically polished down to a thickness of ~120 μm. Thin foils were prepared from these discs by electro polishing in a 25% perchloric acid, 25% ethanol and 50% water electrolyte at room temperature (~22^{o}C), 9 volts and ~20 mA for about 15s.

3 Results

3.1 Microstructure

After each heat treatment, specimens were examined in a transmission electron microscope (TEM) and precipitate size and density were determined. As expected, heat treatments after prime ageing (PA) led to significant coarsening of the prime aged precipitate microstructure. Figure 1 shows the size distributions of precipitates determined after annealing at 600^{o}C for 1, 2 and 4 hours. For comparison, the size distribution for the specimen in the prime aged condition is also shown in Figure 1. The average precipitate size and density for various heat treatments are quoted in Table 1.

Figure 2 shows the size distribution for annealing at 700^{o}C for 4 hours. The annealing at 850^{o}C for 4 hours led to very low density of rather large and heterogeneously distributed precipitates and therefore no quantitative measurements were made. Examples of precipitate microstructure after prime ageing and annealing at 600^{o}C for 4 hours and at 700^{o}C for 4 hours are shown in Figure 3.

Table 1. The average precipitate size and density in CuCrZr alloy after different heat treatments

Heat Treatment	Precipitate Size (nm)	Precipitate Density (m^{-3})
PA*	2.2	2.6×10^{23}
PA + 600°C/1 h	8.7	1.7×10^{22}
“ “ /2 h	9.4	1.8×10^{22}
“ “ /4 h	21.3	1.5×10^{21}
PA + 700°C/4 h	46.4	7.0×10^{20}
PA + 850°C/4 h	NA+)	NA+)

*Prime Aged

+)Precipitate density was too low and too heterogeneous for quantitative measurements

3.2 Tensile properties

The prime aged (PA) and the overaged specimens were tensile tested at 50 and 300°C. All tests were carried out in vacuum ($<10^{-4}$ torr) and at a strain rate of $1.2 \times 10^{-3}\ s^{-1}$. The engineering stress-strain curves for the PA specimen and specimens heat treated at 600°C for 1 and 4 hours tested at 50 and 300°C are shown in Figure 4. Figure 5 shows the stress strain curves for the specimens annealed for 4 hours at 600, 700 and 850°C and tested at 50 and 300°C. For comparison, the stress-strain curves for the prime aged specimens are also shown.

The results shown in Figs. 4 and 5 clearly show that the tensile strength decreases both at 50 and 300°C with increasing annealing time at a given annealing temperature (e.g. 600°C) and with increasing annealing temperature for a given annealing time (i.e. 4 hours). This is consistent with the precipitate microstructure (Table 1) in that as precipitate microstructure coarsens with increasing annealing time and temperature, the tensile strength decreases accordingly.

A number of tensile specimens of CuCrZr alloy in the prime aged condition and specimens heat treated at 600°C for 1 and 4 hours were irradiated at 60 and 300°C to a displacement dose level of ~0.3 dpa. The specimens irradiated at 60°C were tensile tested (in vacuum) at a strain rate of $1.2 \times 10^{-3}\ s^{-1}$ at 22 and 60°C. The specimens irradiated at 300°C to a dose level of 0.3 dpa were tensile tested at 300°C (in vacuum) and also at a strain rate of $1.2 \times 10^{-3}\ s^{-1}$. The engineering stress-strain curves for specimens irradiated at 60 and 300°C are shown in Figs. 6 and 7, respectively. For comparison, stress-strain curves for OFHC-copper irradiated and tested under the same condition as the CuCrZr alloy are also shown in Fig. 6 and 7.

As can be seen in Fig. 6, irradiation at 60°C to a dose level of ~0.3 dpa caused a moderate amount of increase in the yield strength of the prime-aged as well as overaged alloys at 600°C for 1 and 4 hours. The increase in the yield strength is, however, accompanied by a substantial decrease in the ductility of the irradiated CuCrZr alloy with different pre-irradiation heat treatments. What is most significant and important from the technological point of view is the observation that the irradiation at 60^{o}C to 0.3 dpa did not cause any noticeable yield drop and plastic instability. It should be pointed out, however, that although the overageing heat treatments have eliminated the technological problem of plastic instability at the very beginning of the plastic deformation, the irradiated CuCrZr alloy still suffers from the lack of work hardening and the reduction in the ductility. Some further adjustment of the pre-irradiation microstructure by thermo-mechanical treatments, for instance, may be necessary to address these problems.

The results of post-irradiation tensile tests on specimens irradiated and tested at 300^{o}C (Fig. 7) demonstrate that the CuCrZr alloy both in the prime aged and overaged conditions becomes noticeably softer due to irradiation at 300^{o}C to 0.3 dpa.

The measured values of 0.2% engineering stress ($\sigma_{0.2}$), this ultimate tensile stress (σ_{max}) and the uniform and total elongations (ε_u and ε_t) are quoted in Table 2.

Table 2. Effect of heat treatments on tensile properties of CuCrZr alloy before and after irradiation at 60 and 300^{o}C to 0.3 dpa

Test No.	Heat Treatment	Irr. Temp. (^{o}C)	Dose (dpa)	Test Temp. (^{o}C)	$\sigma_{0.2}$ (Mpa)	σ_{max} (Mpa)	ε_u (%)	ε_t (%)
1225	PA	-	-	50	290.0	398.0	24.0	28.0
1226	”	-	-	50	295.0	416.0	22.0	30.0
1382	”	-	-	60	280.0	373.1	21.3	25.4
1384	“	-	-	60	260.0	364.0	20.0	24.0
1217	HT1	-	-	50	200.0	318.0	26.0	30.0
1211	HT2	-	-	“	175.0	289.0	24.0	32.0
1218	“	-	-	“	165.0	307.0	34.0	41.0
1219	PA	-	-	300	240.0	304.0	18.0	27.0
1220	“	-	-	“	250.0	328.0	17.0	23.0
1385	“	-	-	“	244.0	308.0	15.0	19.0
1221	HT1	-	-	300	180.0	255.0	19.0	24.0
1222	“	-	-	“	150.0	227.0	17.0	21.0
1223	HT2	-	-	300	135.0	218.0	22.0	28.0
1224	“	-	-	“	120.0	201.0	25.0	36.0
1360	PA	60	0.3	60	435.0	436.0	2.0	6.0
1353	”	“	“	22	450.0	458.6	1.7	6.8
1354	HT1	“	“	22	410.0	422.2	1.8	9.6
1358	”	“	“	60	397.0	397.0	2.0	15.0
1355	HT2	“	“	22	360.0	363.0	1.4	9.7

1359	”	“	“	60	372.0	372.0	1.0	7.0
1386	PA	300	“	300	210.0	268.0	10.0	12.0
1577	“	“	“	“	190.0	245.0	6.0	8.0
1387	HT1	“	“	300	-	209.0	14.0	18.0
1576	“	“	“	“	134.0	192.0	9.0	12.0
1388	HT2	“	“	300	-	193.0	21.0	27.0
1389	“	“	“	“	119.0	188.0	20.0	26.0

PA: Prime aged.

HT1: PA + 600^{o}C/1 h; HT2: PA + 600^{o}C/4 h.

3.3 Post-deformation microstructure

Microstructures of the CuCrZr alloy with different heat treatments and irradiated at 60 and 300^{o}C to 0.3 dpa were investigated using transmission electron microscopy in the as-irradiated as well as in the irradiated and deformed conditions.

Even though none of the irradiated specimens exhibited any noticeable yield drop and plastic instability, yet the evidence of plastic flow localization in the form of cleared channels was found in all of the irradiated and deformed specimens (see Fig. 8). The localized deformation in the cleared channels may be responsible for the lack of work hardening as well as the reduction in the ductility of specimens irradiated at 60^{o}C to 0.3 dpa.

3.4 Fracture toughness behaviour

The data measured during fracture toughness testing of three point bend specimens, e.g., load, load line displacement and potential drop values are presented in normalised form in order to make it more informative to compare the results of individual tests and material conditions. The normalised load-displacement curves give qualitative information on the effects of heat treatment, test temperature etc. on fracture behaviour of three point bend specimens.

Typical normalised load-displacement curves for CuCrZr alloy in the unirradiated and neutron irradiated conditions are shown in Figure 9. The three point bend load-displacement curves show a general trend which is similar to that exhibited by the tensile stress-strain curves, i.e., decrease in the strength level due to overaging compared to that in prime aged condition. At the ambient temperature the unirradiated CuCrZr alloy shows relatively flat normalised load-displacement curve without any clear maximum indicating an extensive plastic deformation and crack tip blunting without clear crack extension, i.e., the CuCrZr base alloy has relatively high fracture toughness. In the overaged conditions the normalised load-displacement curves are at lower load levels but the curves have a similar general form to that in the prime aged condition. In the neutron irradiated condition the load levels of the normalised load-displacement curves increase and the general form is more flat and goes through a maximum when compared to that in the unirradiated condition at ambient temperature. This indicates that the CuCrZr alloy had relatively high toughness also after irradiation at the ambient temperature. The most pronounced change is observed when the CuCrZr alloy is irradiated and tested at 300°C, i.e., in both prime aged and overaged conditions the normalised load-displacement curves show a clear maximum indicating relatively low fracture toughness.

Fracture resistance curves of CuCrZr alloy in the unirradiated and the neutron irradiated conditions are shown in Figure 10. In the unirradiated condition clear stable crack growth was observed only in the prime aged condition at 200°C where the initiation fracture toughness J_Q value was about 180 kJm^{-2}. In all the other condition strong crack tip blunting was observed and in those cases the maximum J-integral values are reported in Table 3. In the irradiated condition crack tip blunting without any stable crack growth was also observed when irradiation and fracture toughness tests were carried out at the ambient temperature. Relatively low initiation fracture toughness values of about 33 kJm^{-2} and 82 kJm^{-2} were observed in the prime aged and the overaged conditions, respectively, when irradiated and fracture toughness tested at 300°C. It is noted that only those J_Q values for the prime aged CuCrZr alloy which were irradiated and tested at 300°C are valid according to limitations ($J_{max} = b_0\sigma_y/20$, where b_0 is remaining ligament and σ_y is flow stress) set for specimen dimensions. Limiting values of J_{max} for the prime aged and overaged conditions are about 90 and 83 kJm^{-2} at the ambient temperature and 45 and 35 kJm^{-2} at 300°C, respectively.

Table 3. Effect of heat treatments on initiation fracture toughness of CuCrZr alloy before and after irradiation.

Test No.	Heat treatment	Irr. temp. (°C)	Dose (dpa)	Test temp. (°C)	J_Q (kJm^{-2})	
M2562	PA	-	-	100	230	*
M2585	PA	-	-	200	183	
M2565	HT1	-	-	100	224	*
M2586	HT1	-	-	200	285	*
M1993	HT2	-	-	20	220	*
M2566	HT2	-	-	100	200	*
M2873	HT2	-	-	200	154	*
M2804	PA	60	0.3	20	301	*
M2805	PA	60	0.3	20	400	*
M2807	PA	60	0.3	20	416	*
M2806	HT1	60	0.3	20	342	*
M2808	HT1	60	0.3	20	304	*
M2809	HT1	60	0.3	20	348	*

M2821	PA	300	0.3	300	26	
M2822	PA	300	0.3	300	20	
M2859	PA	300	0.3	300	25	
M2858	PA	300	0.3	300	38	
M2857	PA	300	0.3	300	58	
M2860	HT1	300	0.3	300	72	
M2861	HT1	300	0.3	300	84	
M2862	HT1	300	0.3	300	89	

PA: Prime aged.

HT1: PA + 600^{o}C/1 h; HT2: PA + 600^{o}C/4 h.

* crack tip blunting

4 Summary and conclusions

The changes in the precipitate microstructure of the CuCrZr alloy due to different heat treatments were quantitatively characterized using transmission electron microscopy. Results showed that the precipitate density decreases very rapidly with increasing overageing time at 600^{o}C. For instance, the overageing for 2 and 4 hours led to a reduction in precipitate density by a factor of about 10 and 200, respectively, compared to the density in the prime aged condition. The overageing at 850^{o}C for 4 hours caused almost a complete resolution of the precipitates. The reduction in the precipitate density must be responsible for a significant decrease in the tensile strength of the alloy due to overageing. On the other hand overaging did not have any significant effects on fracture toughness properties.

The irradiation at 60^{o}C to a dose level of ~0.3 dpa caused some noticeable amount of hardening both in the prime aged and overaged specimens. The ductility, on the other hand, remained significantly reduced. The irradiation did not caused any noticeable changes in fracture toughness properties at ambient temperature. The irradiation at 300^{o}C to a dose level of ~0.3 dpa did not cause any hardening. However, initiation fracture toughness was significantly reduced in both prime aged and overaged conditions at 300^{o}C.

The most significant effect of the overageing heat treatments was that the heat treatment eliminated the problem of yield drop and plastic instability. However, the overageing heat treatments neither restored the work hardening ability nor the ductility of the alloy. The post-deformation microstructure of the irradiated alloy suggests that even the overaged alloy suffers from the problem of flow localization in the form of cleared channels and this may by responsible for the lack of work hardening as well as the reduction in ductility due to irradiation.

Acknowledgements

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Figures

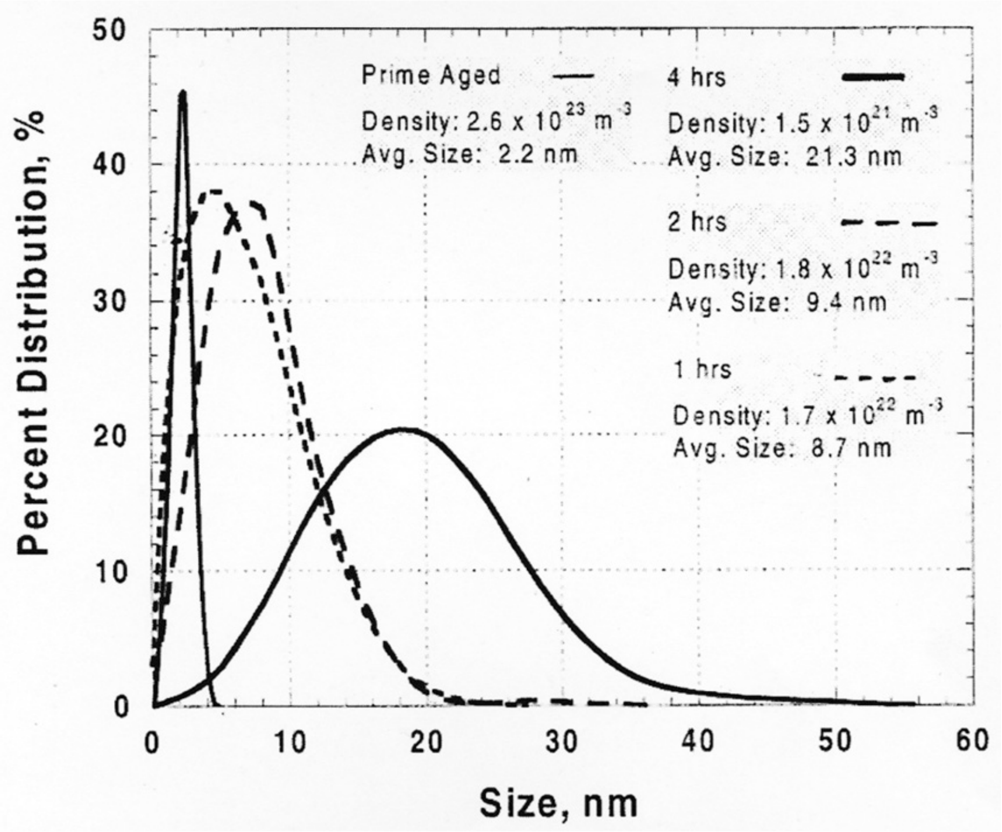


Figure 1. Precipitate size distribution in the unirradiated CuCrZr alloy in the prime aged and heat treated (after prime ageing) specimens at 600°C for 1, 2 and 4 hours.

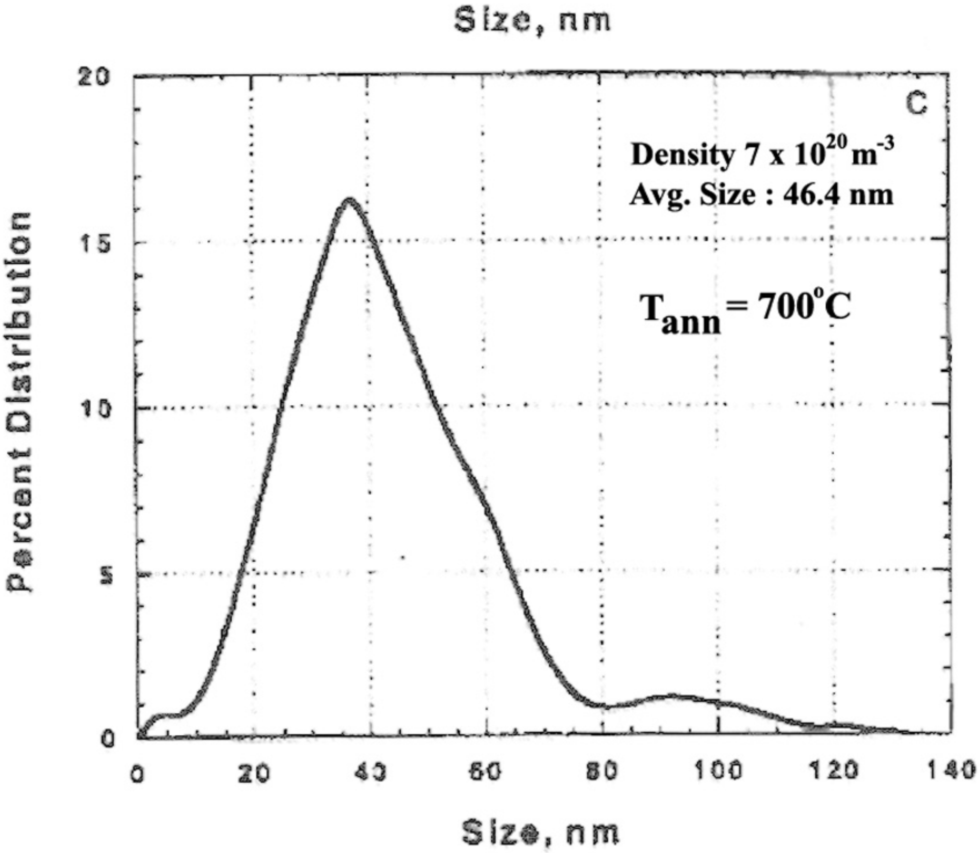


Figure 2. Precipitate size distribution in the unirradiated CuCrZr alloy overaged (after prime ageing) at 700^{o}C for 4 hours.

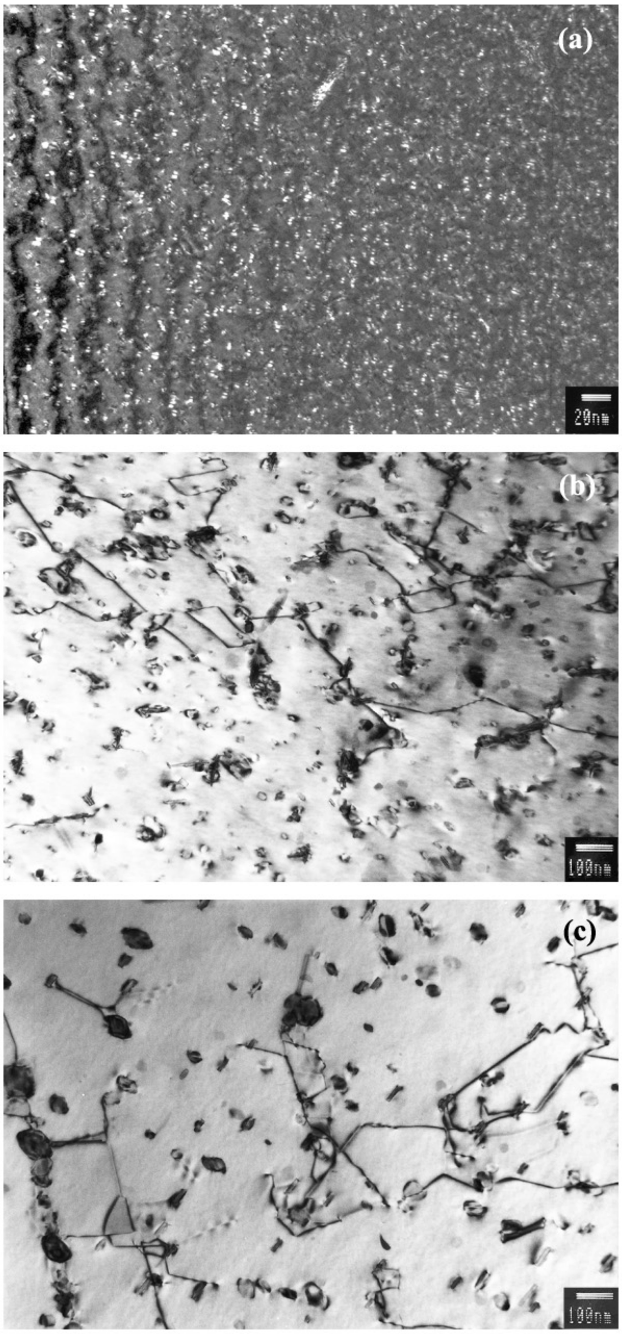


Figure 3. Examples of precipitate microstructure after (a) prime ageing, (b) annealing at 600°C for 4 hours and (c) annealing at 700°C for 4 hours.

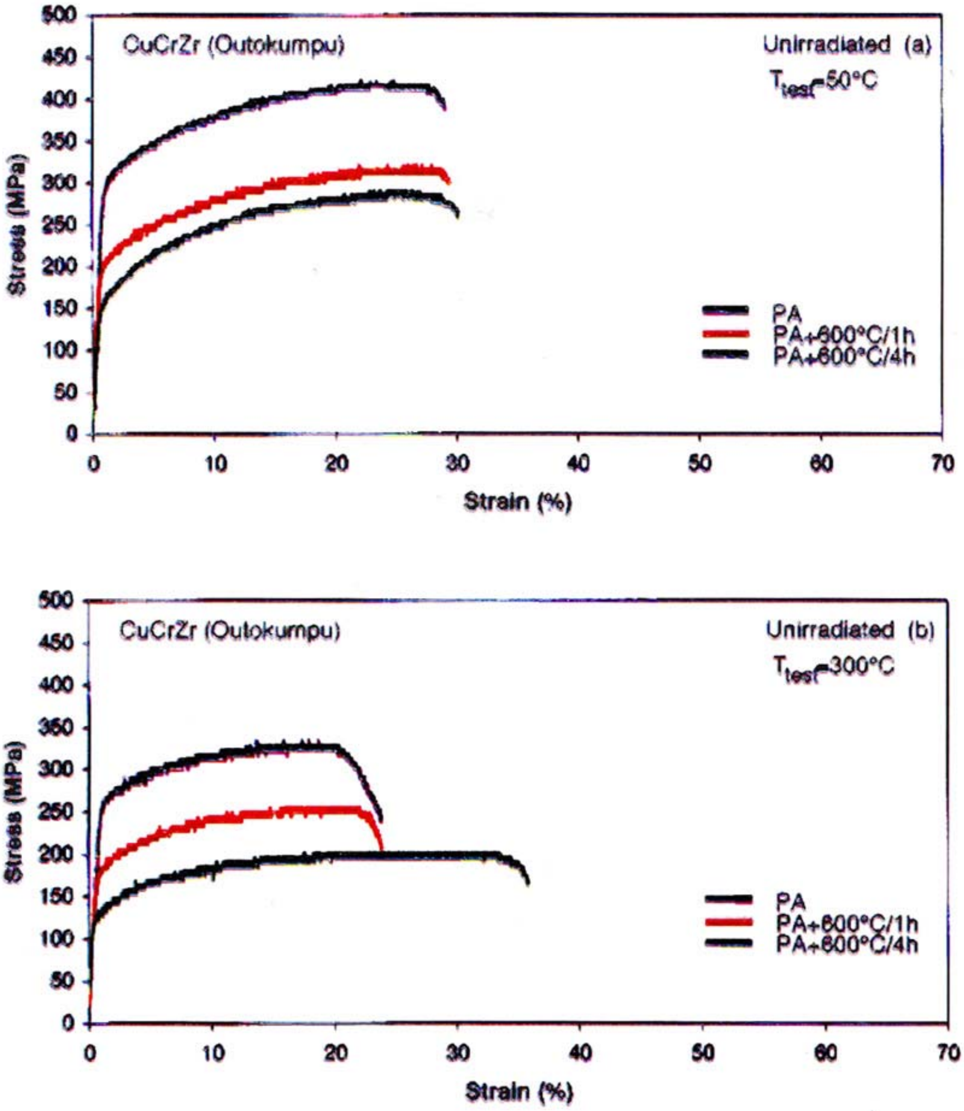


Figure 4. Stress-strain curves for CuCrZr alloy in the prime-aged condition and after annealing at 600°C for 1 and 4 hours and tensile tested at (a) 50°C and (b) 300°C.

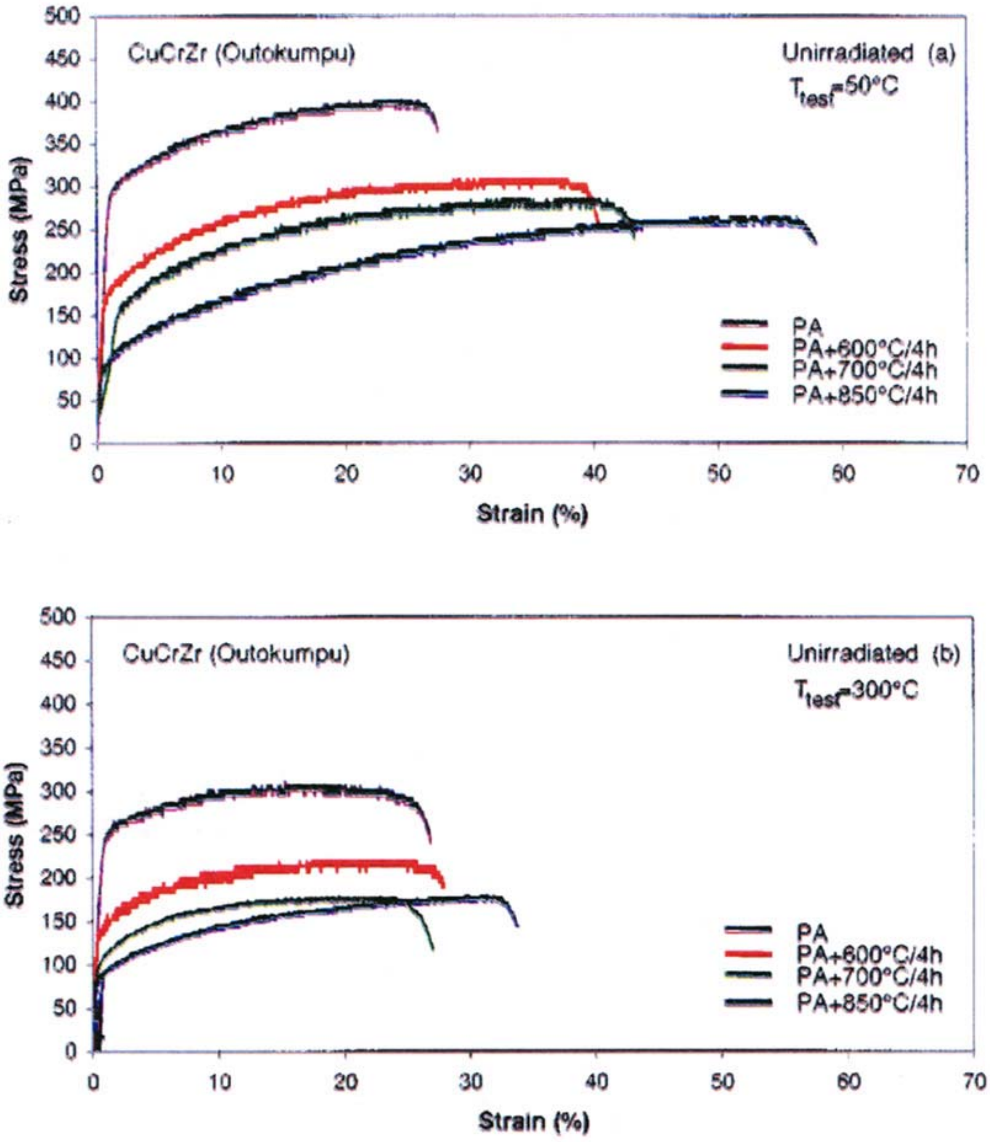


Figure 5. Stress-strain curves for CuCrZr alloy heat treated (after prime ageing) at 600, 700 and 850°C for 4 hours and subsequently tensile tested at (a) 50°C and (b) 300°C.

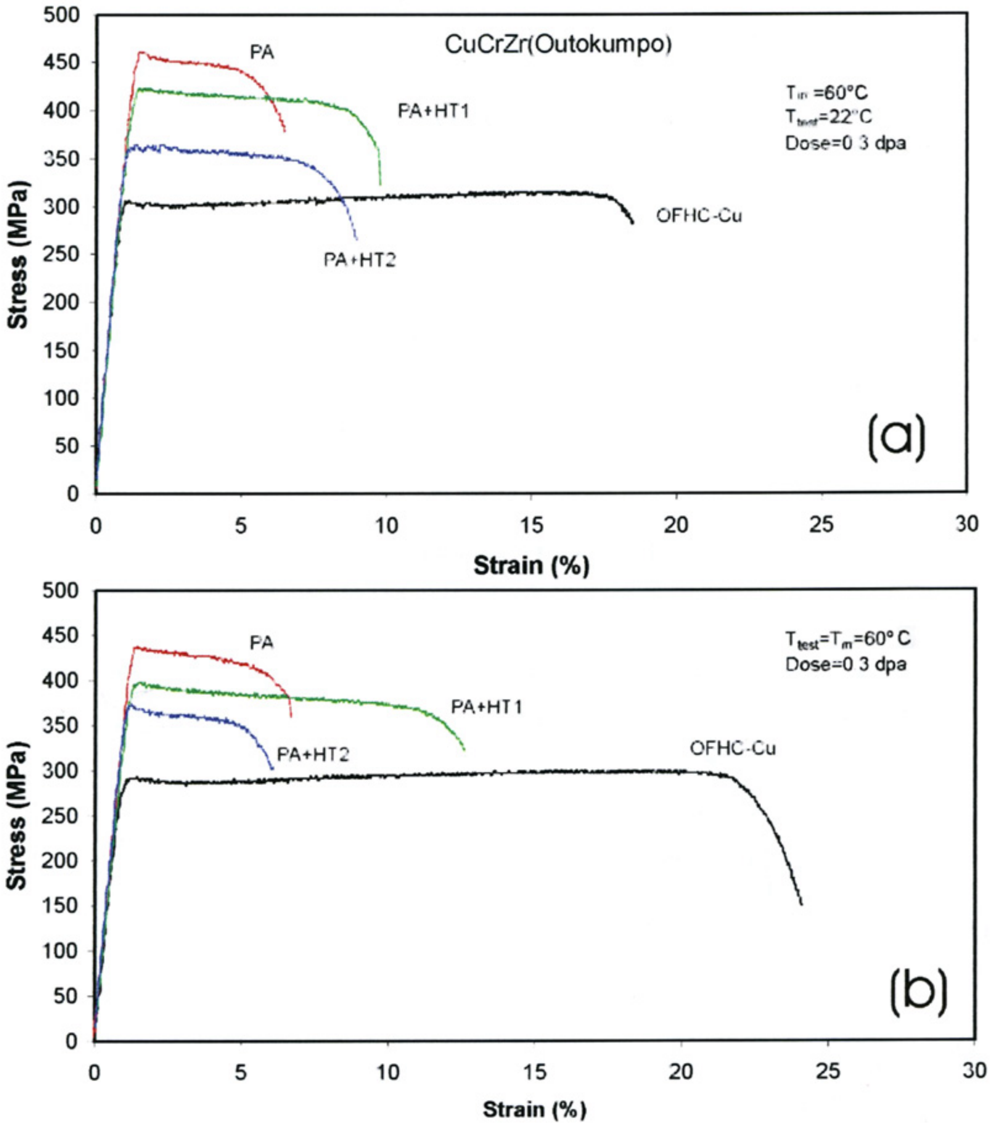


Figure 6. Stress-strain curves for CuCrZr alloy irradiated at 60°C to a dose level of ~0.3 dpa in the prime aged and overaged (at 600°C for 1 h (PA + HT1) and 4 h (PA + HT2)) conditions and tensile tested at (a) 22°C and (b) 60°C.

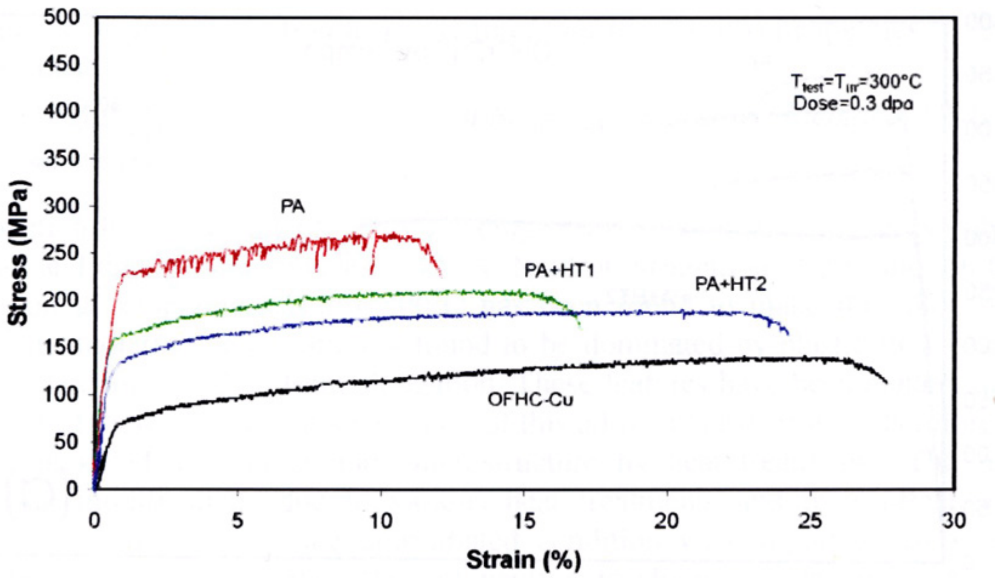


Figure 7. Same as Figure 6 but irradiated and tensile tested at 300°C.

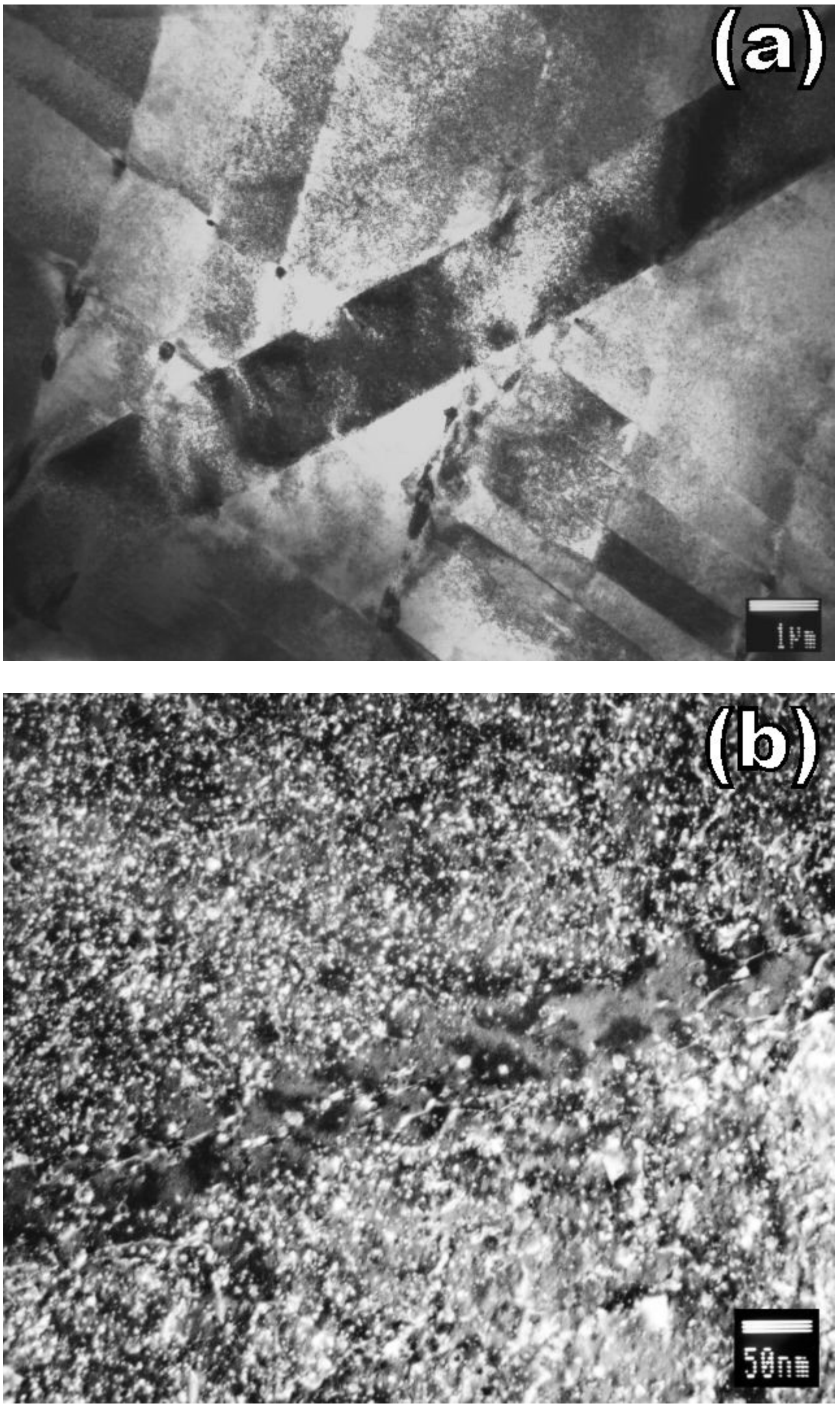


Figure 8. (a,b)

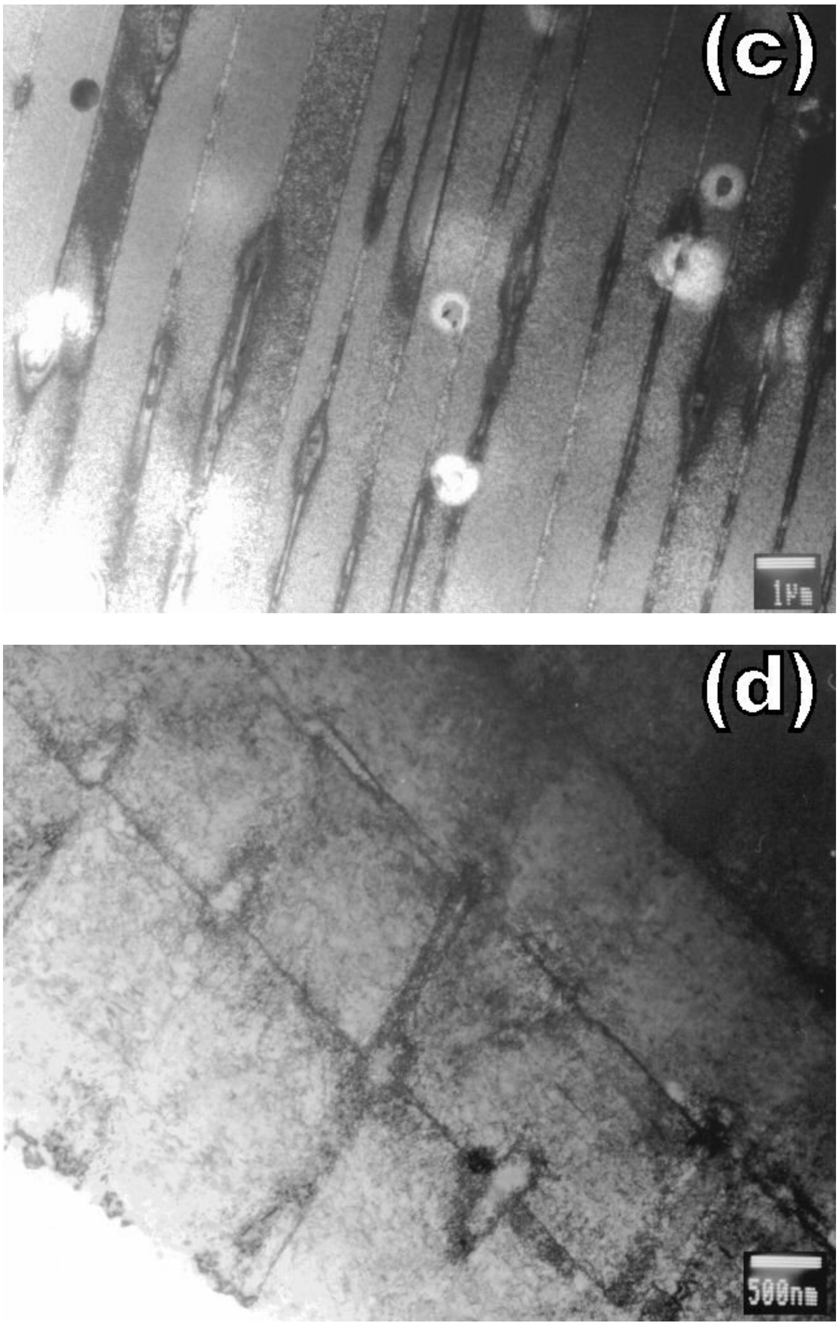


Figure 8. Examples of cleared channel formation in specimens irradiated at 60°C to ~0.3 dpa and deformed at 60°C: (a) prime aged, (b) overaged at 600°C for 1 h and (c,d) overaged at 600°C for 4 hours.

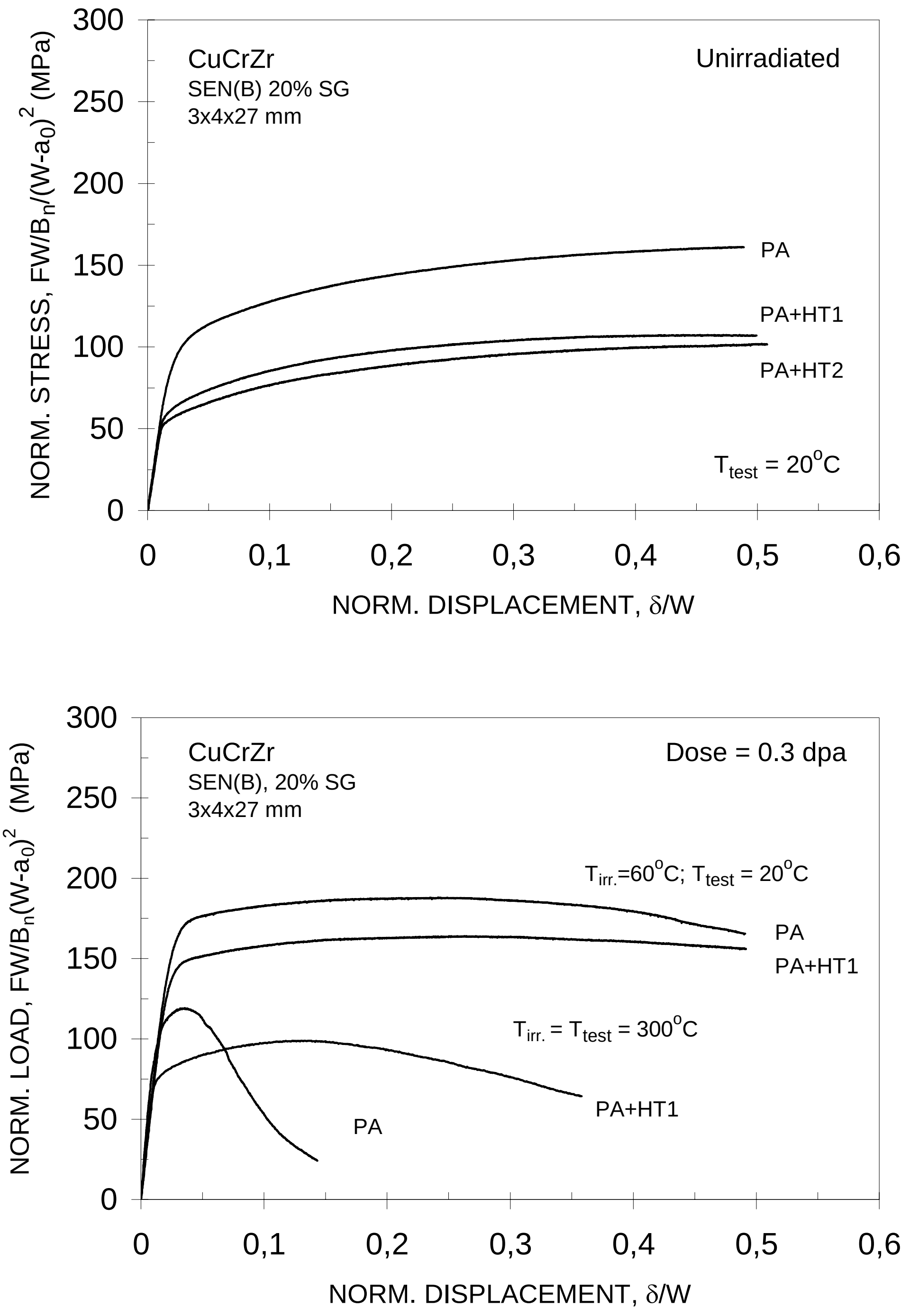
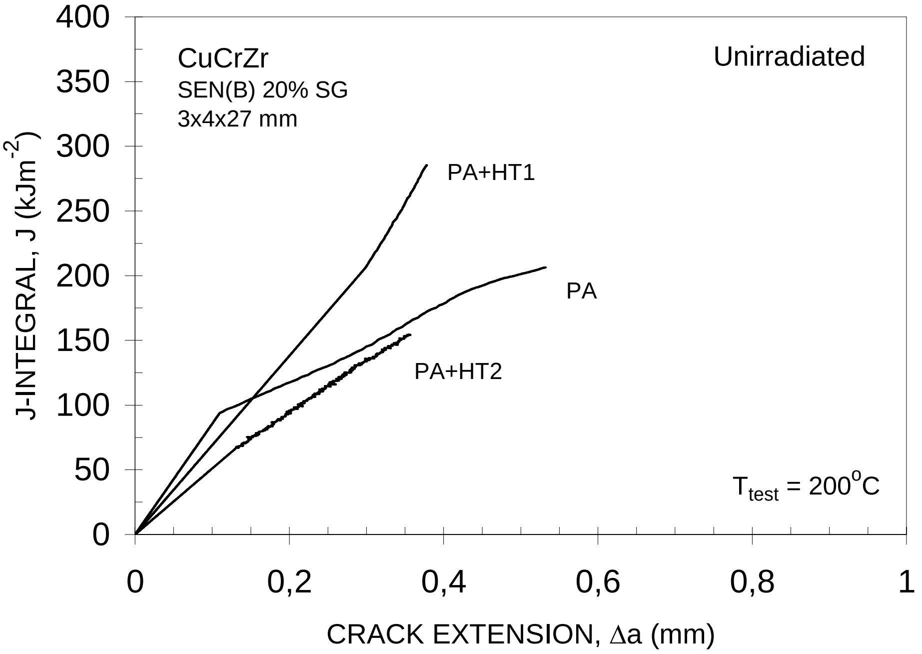


Figure 9. Normalised load-displacement curves for CuCrZr alloy (a) unirradiated in the prime aged and overaged (at 600°C for 1 h (PA + HT1) and 4 h (PA + HT2)) conditions at 20°C and (b) irradiated at 60°C and 300°C and tested at 20°C and 300°C, respectively, to a dose level of ~0.3 dpa in the prime aged and overaged (at 600°C for 1 h (PA + HT1)) conditions.



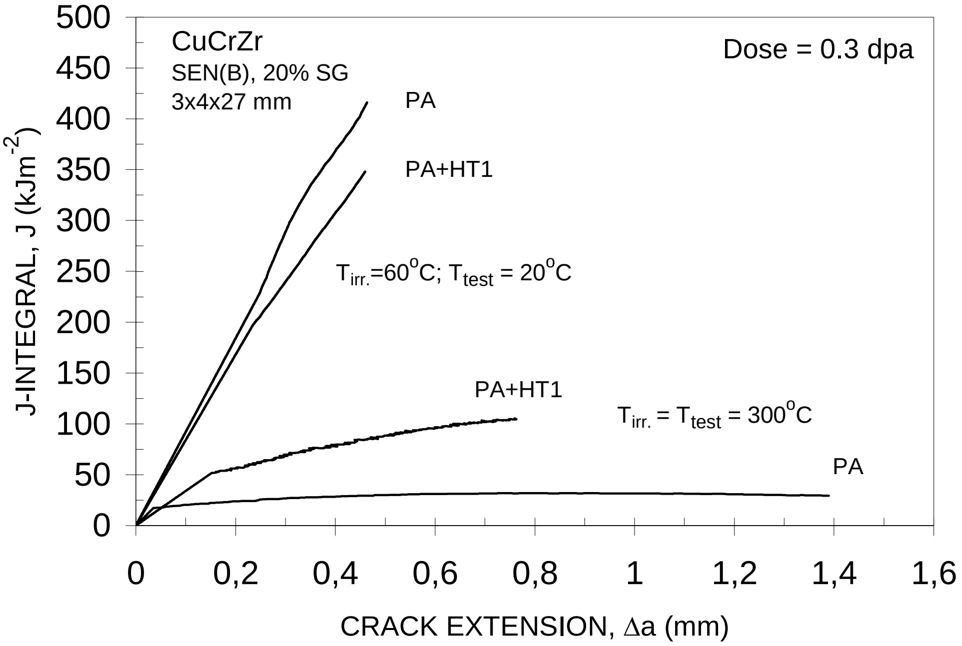


Figure 10. Fracture resistance curves of CuCrZr alloy (a) unirradiated in the prime aged and overaged (at 600°C for 1 h (PA + HT1) and 4 h (PA + HT2)) conditions at 200°C and (b) irradiated at 60°C and 300°C and tested at 20°C and 300°C, respectively, to a dose level of ~0.3 dpa in the prime aged and overaged (at 600°C for 1 h (PA + HT1)) conditions.

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