Grain refinement by utilizing grain-boundary reaction in heat-resistant alloys

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The possibility of grain refinement by thermal cycling is examined on commercial heat-resistant alloys, namely, an austenitic 21Cr–4Ni–9Mn steel and a cobalt-base HS-21 alloy, in which the grain-boundary reaction occurs. A thermal cycle is composed of a high-temperature ageing which causes the grain-boundary reaction, and a subsequent short-term heating at the resolution temperature, which ensures complete dissolution of the grain-boundary reaction precipitates into the matrix. The grain diameter is finally reduced to about one-half or one-third of the original grain size after four thermal cycles, while a larger grain-size reduction is observed in the specimens with initially larger grain size. The effects of the amount of the grain-boundary reaction and the heat-treatment conditions on grain refinement are also experimentally discussed.

1. Introduction

Grain refinement in steels is usually accomplished by a thermomechanical heat treatment such as controlled rolling [1–3], and leads to an increase in yield stress and a lowering of the impact transition temperature. Grain refinement can also be achieved by thermal cycling which utilizes phase transformation in steels [1, 2, 4], although this procedure is not applicable to alloys that do not undergo phase transformation. Scharfenberger *et al.* [5] have proposed a new procedure of grain refinement by using the grain-boundary reaction in a copper–silver alloy. This procedure is also applicable to commercial alloys in which cellular precipitates containing carbide phase are formed by the grain-boundary reaction, but a possibility of grain refinement by utilizing the grain-boundary reaction precipitates has not been studied in these alloys.

In this study, grain refinement by utilizing the grain-boundary reaction precipitates, was examined on two commercial heat-resistant alloys, namely, an austenitic 21Cr–4Ni–9Mn steel and a cobalt-base HS-21 alloy. The effects of the amount of the grain-boundary reaction and the heat-treatment conditions on grain refinement in these alloys were also experimentally discussed.

2. Experimental procedure

Table I shows the chemical composition of the alloys used in this study. Disc-shaped specimens of about 5 mm thickness were cut from the hot-forged bars of 16 mm diameter. Specimens were initially solution-heated for 3.6 or 14.4 ks in the temperature range 1473–1573 K to obtain specimens of a certain grain size, and were then furnace-cooled to 1173 K. Specimens were further aged at 1173 K to develop various amounts of grain-boundary reaction precipitates, because the grain-boundary reaction in these alloys predominantly occurs at 1173 K [6–9].

Subsized specimens for grain refinement of about 5 mm × 5 mm × 5 mm, were cut from the above specimens. The subsized specimens enabled rapid heating to the resolution temperature within 60 s. Fig. 1 shows the thermal cycling procedure for grain refinement of these alloys. Table II shows the heat-treatment conditions for grain refinement employed in this study. A high resolution temperature was chosen which ensured the complete dissolution of precipitates into the matrix during short-term heating (180 or 600 s). Furnace-cooling after resolution-heating was employed to avoid the general precipitation which prevented the formation of grain-boundary reaction precipitates during subsequent ageing at 1173 K. The cooling rate in furnace-cooling was about 0.3 $K s^{-1}$ in the temperature range 1300–1173 K for 21Cr–4Ni–9Mn steel and about 0.05 $K s^{-1}$ for HS-21 alloy [9]. Each thermal cycle was terminated by air-cooling of specimens to room temperature (about 293 K) at the cooling rate of about 1.5 $K s^{-1}$.

The heat-treated specimens were observed with an optical microscope. The average grain diameter of a specimen was obtained by Fullman's method [10] in which the number of grains per unit area was measured on polished surfaces by using an optical microscope. This method may give a larger value of the grain size compared with the mean intercept length grain size [11, 12], because the latter grain size was about 99 μm for specimens of 21Cr–4Ni–9Mn steel solution-heated for 3.6 ks at 1473 K [13]. The amount of grain-boundary reaction (%GBR) (the extent of the grain-boundary reaction precipitates) in the aged specimens was examined by linear analysis as the area fraction of the precipitates [6, 11], using several optical micrographs taken at low magnifications. The

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TABLE I Chemical composition of the alloys used in this study (wt%)

Alloys	C	N	Cr	Ni	Co	Fe	Mn	Mo	B	Si	S	P
21Cr–4Ni–9Mn steel	0.54	0.40	21.10	4.07	–	bal.	9.74	–	–	0.19	0.008	0.017
HS-21 alloy	0.27	–	26.71	2.37	bal.	0.09	0.64	5.42	0.003	0.59	0.007	< 0.005

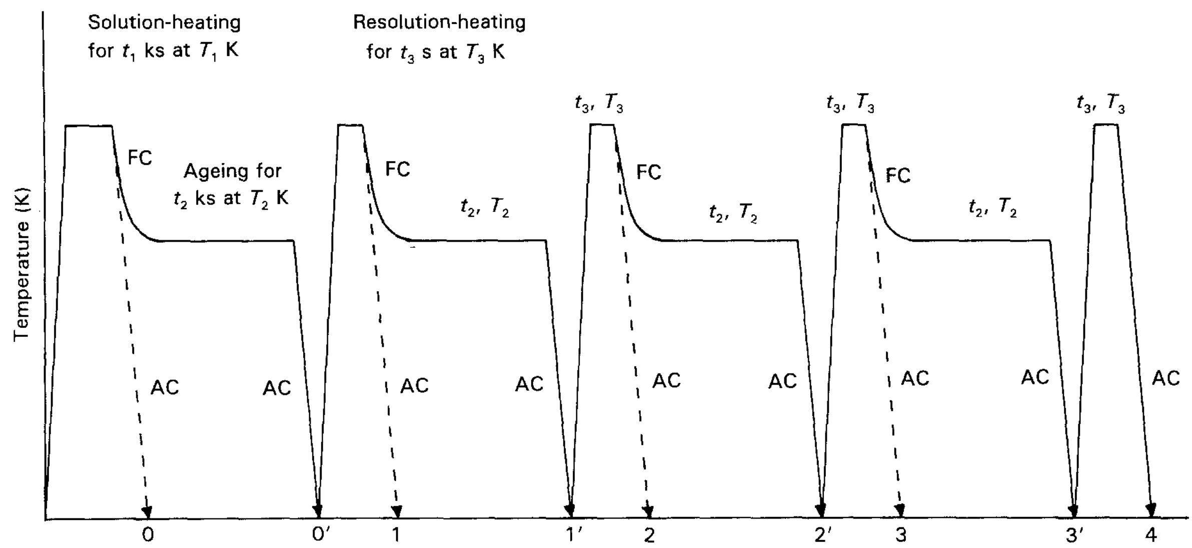

Figure 1 Schematic illustration of the thermal cycling procedure for the grain refinement of the alloys in this study (AC, air-cooled; FC, furnace-cooled).

TABLE II Heat-treatment conditions for grain refinement employed in this study

Alloys	Initial grain diameter (μm)	Solution heating		Ageing condition		Resolution heating	
		t_1 (ks) at	T_1 (K)	t_2 (ks) at	T_2 (K)	t_3 (s) at	T_3 (K)
21Cr–4Ni–9Mn steel	129	3.6	1473	3.6 or 108	1173	180 or 600	1473
	321	3.6	1573	10.8	1173	180 or 600	1473
HS-21 alloy	284	14.4	1523	10.8	1173	180 or 600	1523

extent of the grain-boundary reaction precipitates was also confirmed after ageing at 1173 K in each cycle.

3. Results and discussion

3.1. Grain-boundary reaction

Fig. 2 shows the relationship between the amount of grain-boundary reaction (GBR) and ageing time at 1173 K in specimens furnace-cooled to 1173 K after solution-heating in 21Cr–4Ni–9Mn steel and HS-21 alloy. The amount of GBR increased with ageing time and reached about 100% GBR at 108 ks in the specimen solution-heated at 1473 K (average grain diameter = 129 μm) and at 1080 ks in the specimen solution-heated at 1573 K (average grain diameter = 321 μm) in 21Cr–4Ni–9Mn steel, while the amount of GBR reached at most, about 50% after 1080 ks ageing at 1173 K in HS-21 alloy. The grain-boundary reaction occured more rapidly in specimens with the finer grain size in 21Cr–4Ni–9Mn steel. Fig. 3 shows optical micrographs of the specimens of 21Cr–4Ni–9Mn steel and HS-21 alloy aged at 1173 K. Pearlite-like nodules of the grain-boundary reaction precipitates can be observed in the grains in which no general precipitation, except a small amount of residual precipitates, is visible (Fig. 3a, c and d). It was found in these alloys that the grain-boundary reaction precipitates were composed of rod-like $M_{23}C_6$ precipitates and austenite matrix [6–9]. The amount of GBR reached about 100% after 108 ks ageing in 21Cr–4Ni–9Mn steel (Fig. 3b), while it increased little with time in excess of 10.8 ks in HS-21 alloy.

3.2. Effects of the amount of GBR on grain refinement

Fig. 4 shows the results of grain refinement in the specimens with an initial grain diameter of 129 μm by thermal cycling in 21Cr–4Ni–9Mn steel. At first an ageing time of 108 ks was chosen to cause about 100% GBR in ageing at 1173 K (Fig. 4a). The grain diameter of the specimen decreased from 129 μm to below 70 μm after four thermal cycles, while the shorter heating time at 1473 K for resolution treatment (180 s) gave slightly smaller grain sizes than the longer one (600 s) at each thermal cycle. It is not beneficial to spend much time in heat treatments in the grain refinement of commercial alloys. However, the

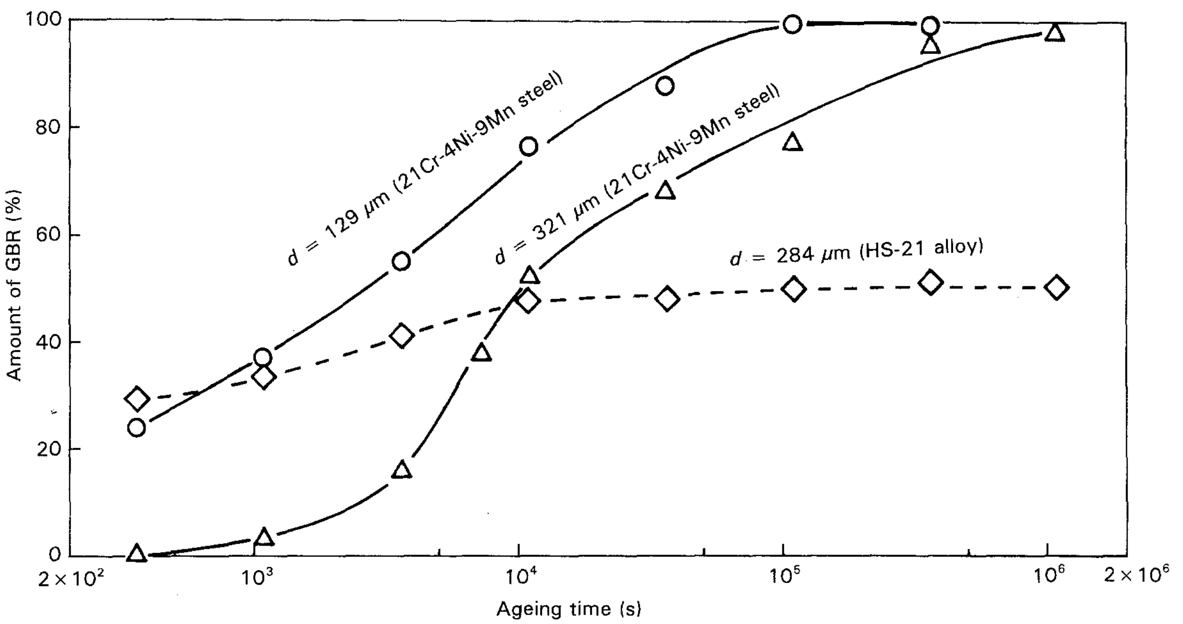

Figure 2 The relationship between the amount of the grain-boundary reaction (GBR) and ageing time at 1173 K in specimens furnace-cooled at 1173 K after solution heating in 21Cr–4Ni–9Mn steel and HS-21 alloy.

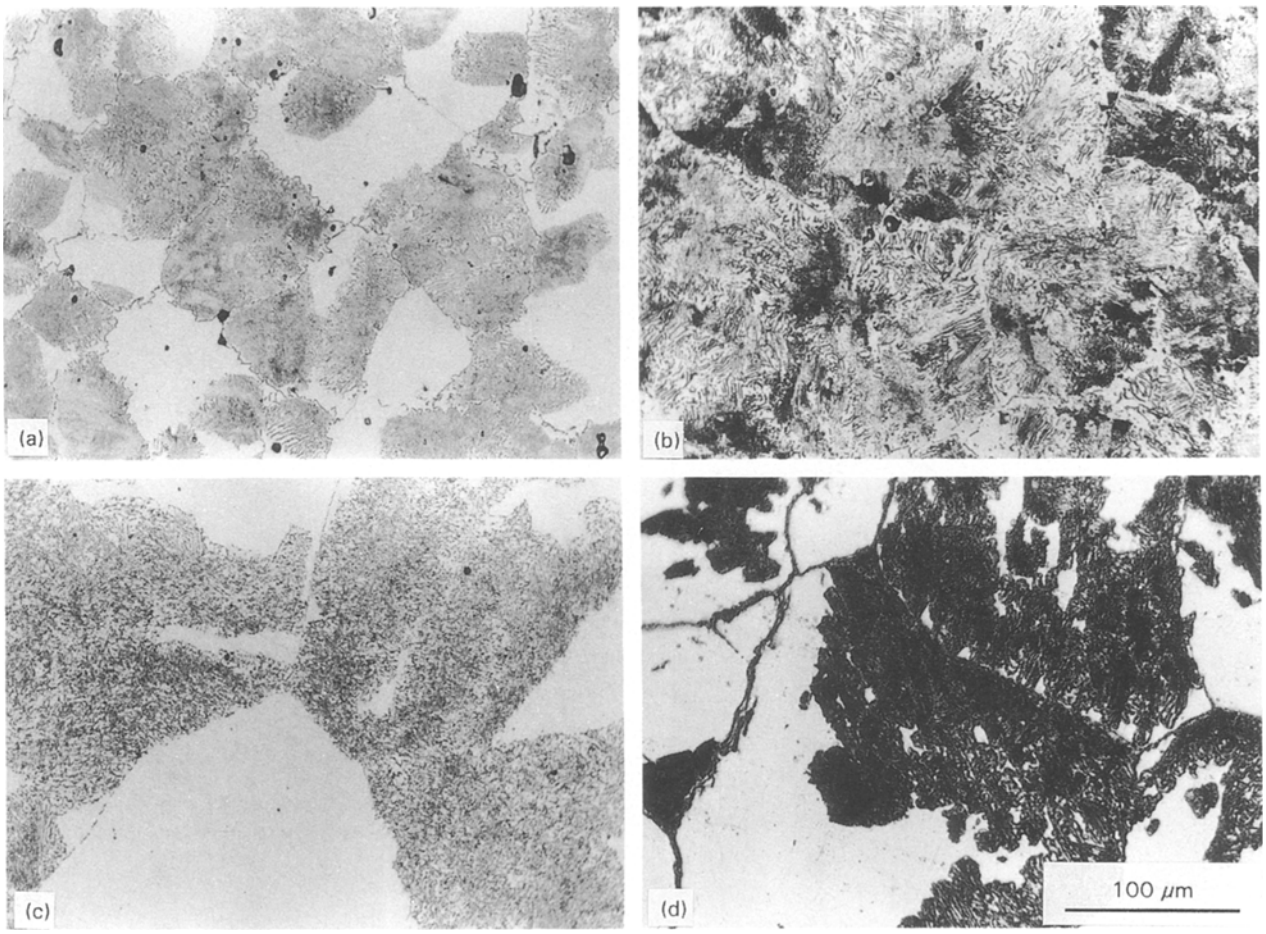

Figure 3 Optical micrographs of the specimens of 21Cr–4Ni–9Mn steel and HS-21 alloy aged at 1173 K. (a) 3.6 ks (d = 129 μm, 55 %GBR, 21Cr–4Ni–9Mn steel), (b) 108 ks (d = 129 μm, 99 %GBR, 21Cr–4Ni–9Mn steel), (c) 10.8 ks (d = 321 μm, 52 %GBR, 21Cr–4Ni–9Mn steel), (d) 10.8 ks (d = 284 μm, 48 %GBR, HS-21 alloy); d = grain diameter, %GBR = amount of grain-boundary reaction.

preliminary study revealed that at least 50% GBR is required to accomplish grain refinement in the 21Cr–4Ni–9Mn steel without causing (fine-grain and coarse-grain) mixed grain structure.

An ageing time of 3.6 ks at 1173 K, which caused about 50% GBR in the specimen with an average grain diameter of 129 μm, was also chosen in the thermal cycling for grain refinement (Fig. 4b). The

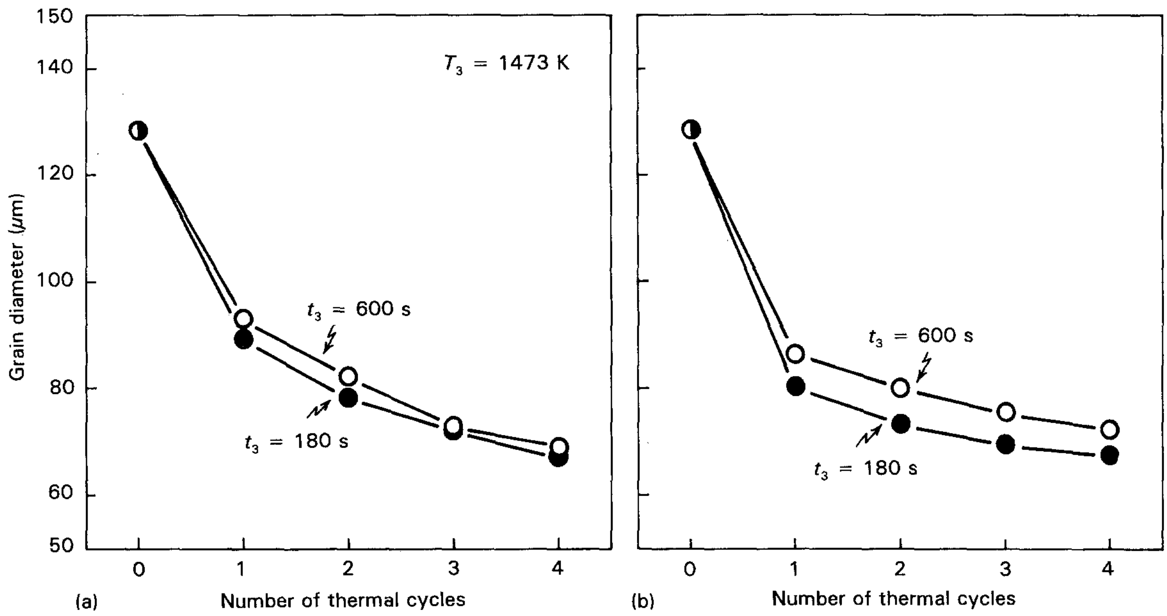

Figure 4 Grain refinement in the specimens with an initial grain diameter of 129 μm by thermal cycling in 21Cr–4Ni–9Mn steel. (a) $t_2 = 108$ ks, $T_2 = 1173$ K; (b) $t_2 = 3.6$ ks, $T_2 = 1173$ K.

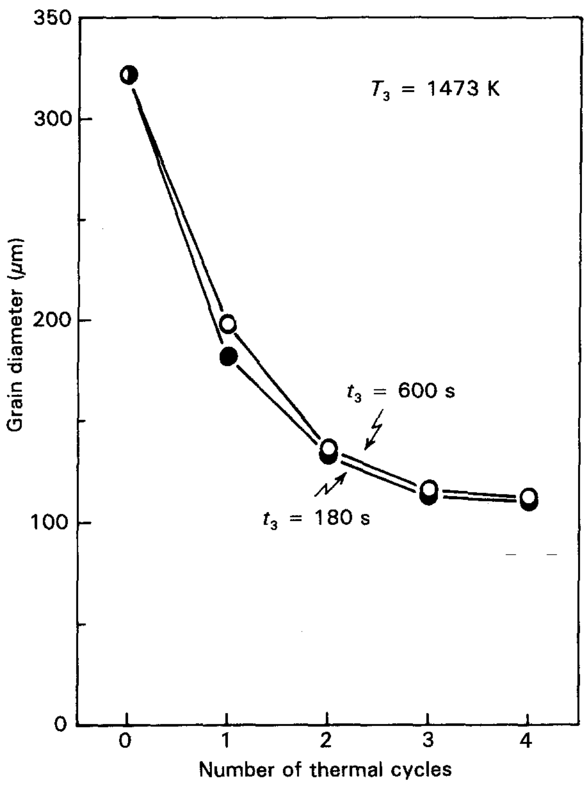

Figure 5 Grain refinement in the specimens with an initial grain diameter of 321 μm by thermal cycling in 21Cr–4Ni–9Mn steel ($t_2 = 10.8$ ks, $T_2 = 1173$ K).

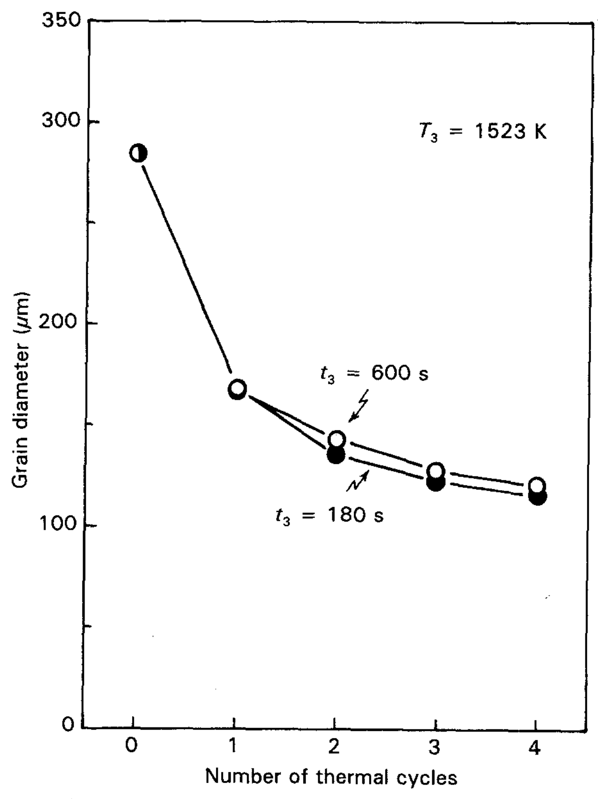

Figure 6 Grain refinement in the specimens with an initial grain diameter of 284 μm by thermal cycling in the cobalt-base HS-21 alloy ($t_2 = 10.8$ ks, $T_2 = 1173$ K).

average grain diameter was reduced to about one-half of the original grain size after four thermal cycles, and the shorter heating at 1473 K gave slightly smaller grain sizes, but the final grain diameter of these specimens was almost the same as that of specimens aged for 108 ks at 1173 K. Thus, it was found in this study that the complete grain-boundary reaction (100% GBR) was not always necessary for grain refinement, in contrast to the results in a copper–silver alloy [5].

3.3. Grain-refinement in coarse-grained specimens

Fig. 5 shows the results of the grain refinement in the specimens with an initial grain diameter of 321 μm in 21Cr–4Ni–9Mn steel. The ageing time of 10.8 ks which caused about 50% GBR in specimens with the original grain size (321 μm) on ageing at 1173 K, was employed in the thermal cycles. The grain diameter decreased from 321 μm to about 110 μm after four

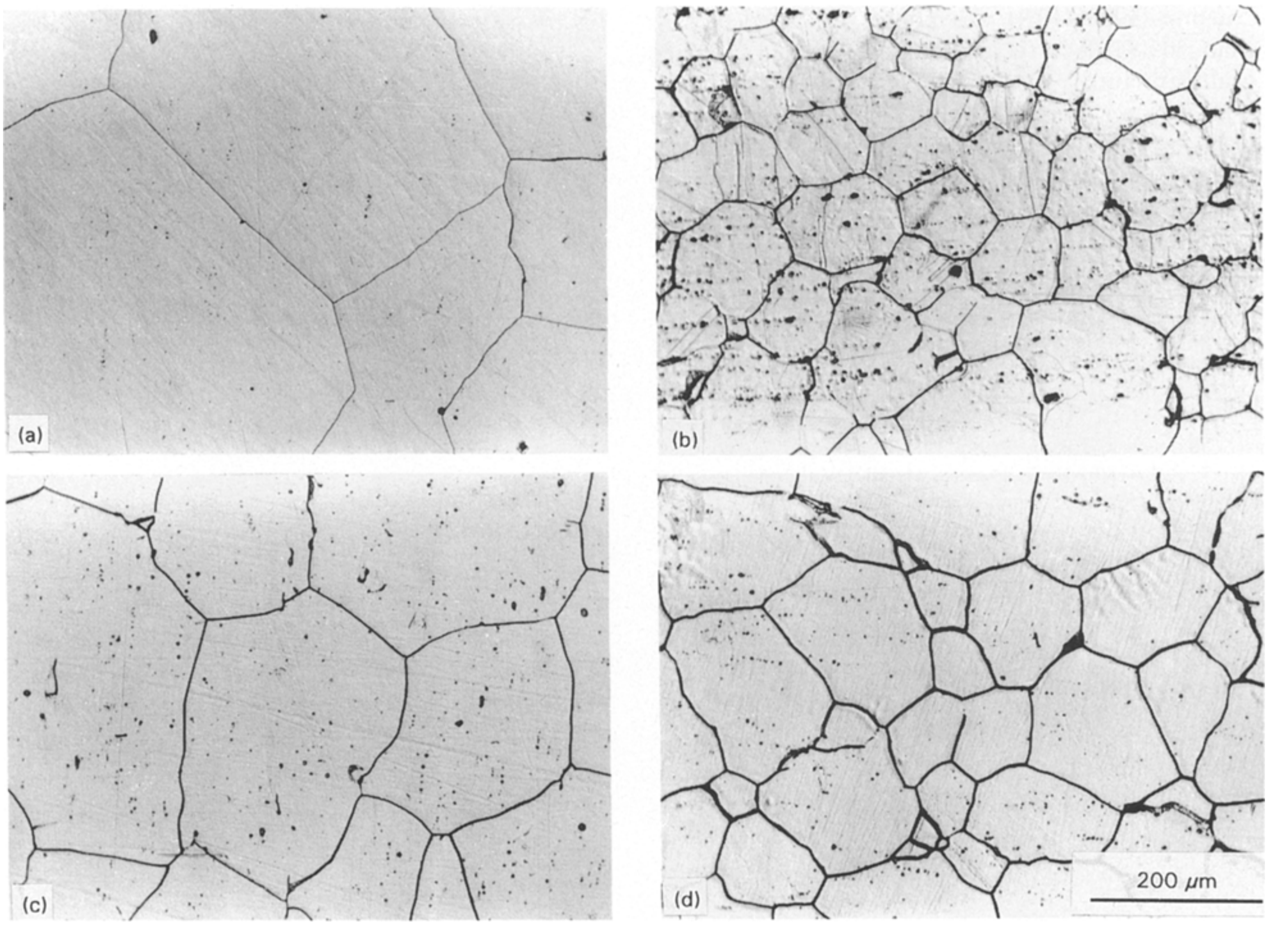

Figure 7 Microstructures of the solution-treated specimens and of the specimens after grain refinement in 21Cr–4Ni–9Mn steel and HS-21 alloy. (a) As solution-treated ($d = 321$ μm, $t_1 = 3.6$ ks, $T_1 = 1573$ K, 21Cr–4Ni–9Mn steel); (b) after four thermal cycles as in (a) ($d = 112$ μm, $t_2 = 10.8$ ks, $T_2 = 1173$ K, $t_3 = 180$ s, $T_3 = 1473$ K, 21Cr–4Ni–9Mn steel); (c) as solution-treated ($d = 284$ μm, $t_1 = 14.4$ ks, $T_1 = 1523$ K, HS-21 alloy); (d) after four thermal cycles as in (c) ($d = 116$ μm, $t_2 = 10.8$ ks, $T_2 = 1173$ K, $t_3 = 180$ s, $T_3 = 1523$ K, HS-21 alloy); d = grain diameter.

thermal cycles. The final grain size was slightly smaller in specimens resolution-heated for the shorter period (180 s). The grain size of the specimens was reduced to about one-third of the original size by the grain refinement in this study. Thus, a larger grain-size reduction by thermal cycling was obtained in specimens with an initially larger grain size in 21Cr–4Ni–9Mn steel. Fig. 6 shows the results of the grain refinement in the specimens with an initial grain diameter of 284 μm in cobalt-base HS-21 alloy. The grain diameter decreased with increasing number of thermal cycles, and finally decreased to about 120 μm after four thermal cycles, although the final grain size was slightly smaller in the specimens resolution-treated for 180 ks at 1523 K.

Fig. 7 shows the microstructures of the solution-treated specimens and of the specimens after grain refinement in 21Cr–4Ni–9Mn steel and HS-21 alloy. There were no precipitates either in the grains or on the grain boundaries in the heat-treated specimens of 21Cr–4Ni–9Mn steel after four thermal cycles (Fig. 7b), while almost no precipitates were visible in the solution-treated specimens of the original grain size (Fig. 7a). The same results were obtained in specimens of HS-21 alloy (Fig. 7c and d). The grain refinement in these alloys was accomplished by a combination of short-term ageing at 1173 K and resolution treatment at 1473 or 1523 K, which ensured the complete dissolution of the precipitates into the austenite matrix, although the grain-size reduction by thermal cycling was smaller in these alloys than in carbon steels [1, 4, 14]. Therefore, this procedure is applicable to the grain refinement of alloys in which a large amount of grain-boundary reaction occurs in the high-temperature ageing.

4. Conclusions

Grain refinement by thermal cycling was examined on 21Cr–4Ni–9Mn steel and HS-21 alloy, in which the grain-boundary reaction occurred. The following results were obtained.

1. A thermal cycle was composed of a high-temperature ageing which caused the grain-boundary reaction, and a subsequent short-term resolutioning treatment which ensured complete dissolution of the grain-boundary reaction precipitates into austenite matrix. The grain diameter finally reduced to about one-half or about one-third of the original grain size after four thermal cycles in these alloys.

2. At least about 50% of the grain-boundary reaction (in area fraction) in the ageing treatment of the thermal cycles was necessary for the grain refinement in these alloys, although the complete grain-boundary

reaction (100%GBR) was not always needed. Less than 50%GBR in the ageing treatment led to mixed grain structure.

3. A larger grain-size reduction by thermal cycling was observed in specimens with an initially larger grain size in 21Cr–4Ni–9Mn steel. The final grain size was slightly smaller in specimens resolution-heated for a shorter time.

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