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Effect of order–disorder transformation modes on the anomalous yield behaviour of Fe_3Al intermetallic compounds

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Compression tests were performed to clarify the effects of transformation modes on the anomalous yield behaviour of hypo- and hyper-stoichiometric Fe_3Al alloys which show a first- and a second-order transition, respectively. There were great differences in the anomalous yield behaviour depending on the transformation modes. In the first-order transformation alloy, changes in the degree of order played an important role before phase separation, while precipitation of α phase had a great influence on the anomalous behaviour after phase separation. In contrast, only the change in the degree of order was a dominant factor in the second-order transformation alloy.

1. Introduction

Iron aluminides near stoichiometric Fe_3Al composition exhibit an increase in yield stress with increasing temperature in an intermediate temperature region, which is very attractive for high-temperature structural application [1]. Much work has been devoted to understanding the exact mechanism of the anomalous increase in yield stress with temperature.

Stoloff and Davies [2] explained the anomalous yield behaviour by the generation of antiphase boundary (APB) during transition from superdislocation to unit dislocation near the order–disorder transformation temperature. Hanada *et al.* [3] referred the anomalous behaviour to thermally activated cross-slip pinning mechanism in addition to the Stoloff–Davies mechanism. Recently, however, Inouye [4] observed that α precipitation and B2 formation are responsible for the positive temperature dependence of yield stress. Although Stoloff and Davies' model seems not to be applicable to the age-hardenable alloy, Inouye's experiments do not clearly show how strengthening by B2 formation is possible in an alloy which has no α precipitation at all.

Order–disorder transformations in Fe_3Al intermetallic compounds take place by a continuous process and/or a nucleation-growth process [5]. The widely accepted phase diagram near Fe_3Al composition is presented in Fig. 1 [6]. Phase transformation in a hypo-stoichiometric composition alloy, which is a first-order process in nature according to the Ehrenfest classification, can occur by the parallel reaction of continuous process and nucleation-growth process, while equilibrium transformation in hyper-stoichiometric composition, which is a second-order process, occurs by the continuous process alone [5, 7]. This may lead to different strengthening, and therefore, different effects on the anomalous behaviour.

The purpose of the present work was to investigate the differences in the anomalous yield behaviour between hypo- and hyper-stoichiometric Fe_3Al alloys, and to elucidate the dominant factor responsible for the anomalous behaviour.

2. Experimental procedure

Alloys containing 24.1% and 27.2% Al (at % only in the case of Fe–Al alloy) were made from 99.95% electrolytic iron and 99.9% aluminium by vacuum induction melting and cast into steel moulds as 60 mm × 60 mm × 130 mm ingots. The ingots were hot forged and swaged to about 8 mm diameter rods starting at 1100 °C and finishing at 700 °C. Some of the ingots were hot forged and rolled to 1.4 mm thick plate in the same temperature range for microstructural analysis.

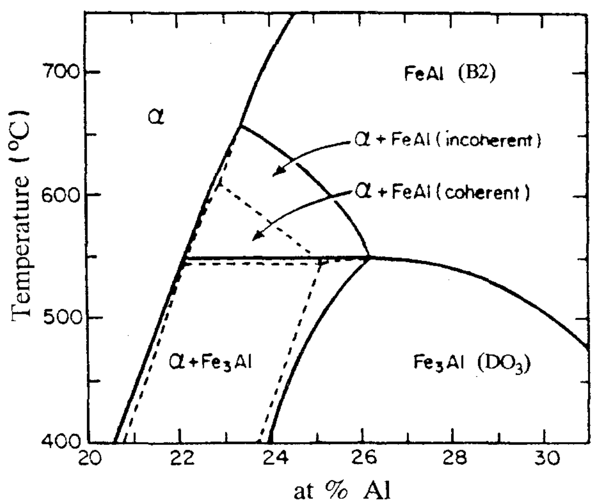

Figure 1 Fe–Al phase diagram near Fe_3Al [6].

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Compression specimens with diameter 6 mm and length 9 mm were machined from the round bars.

The specimens encapsuled in an evacuated quartz tube were annealed at 800 °C for 30 min in a resistance heating furnace, followed by furnace cooling at a rate of 5 °C^{-1} and DO_3 ordering at 400 °C for 50 h and then quenching into water. Ageing was subsequently done in the temperature range 480–660 °C at an interval of 30 °C for times ranging from 0.1–1000 min. Ageings for 100 min or less were done on the compression testing machine after rapid heating at a rate of 50 °C sec^{-1}, while ageings for 1000 min were conducted in the resistance furnace. Mean diameters of grain size after heat treatment were in the range 60–80 μm.

Compression tests were performed on the Thermecmastor-Z which had been developed for precisely simulating various deformation processes using computer systems, high-vacuum systems, electro-hydraulic servo systems, laser measuring systems and induction heating systems for thermocouple-welded specimens. Deformation of aged specimens was carried out at each ageing temperature at a strain rate of 0.01 s^{-1} for the purpose of maintaining as long as possible the degree of order during deformation. The true stress–strain curves were obtained by computer-aided automatic data processing, and 0.2% offset yield stresses were obtained from them.

Specimens for transmission electron microscopy (TEM) were cut into 10 mm squares from the rolled plates and ground to 0.3 mm thickness. The thin plates were DO_3 ordered with the same process mentioned above, and then heat treated in a salt bath for 0.1, 10 and 30 min ageing and in the resistance furnace for 100 min ageing. All the specimens were quenched into agitated iced brine. Thermocouple-embedded measurement showed that it had taken about 2–3 s for the samples to reach the desired temperature in the salt bath. The initial heating rate was so fast that most of the increase in time occurred while heating above two-thirds of the ageing temperature, which was quite reasonable to simulate the process of the ageing treatment in the Thermecmastor-Z.

Thin foils for TEM were prepared by mechanical polishing to 50–100 μm thickness and twin jet polishing in 30% nital electrolyte with a current density of 1 A cm^{-2}. Electron microscopy was performed in JEM 200CX electron microscope operating at 200 kV. The type of APBs and phases were determined from the relations between the superlattice reflection and corresponding micrographs which had been proposed by Marcinkowski and Brown [8].

3. Results

The effect of ageing time on the temperature dependence of 0.2% offset yield stress for the 24.1% Al alloys is shown in Fig. 2. In all cases, a distinct increase in yield stress appears with increasing temperature. As the ageing time increases, the positive temperature dependence region shifts to the lower side of temperature. According to the phase diagram of the Fe–Al system shown in Fig. 1, the positive temperature dependence regions correspond to $\alpha + DO_3$ two-phase field. Because the initial state of the alloy is DO_3 phase, α precipitation during ageing in the two-phase region may contribute to the anomalous temperature dependence of yield stress.

Another feature in Fig. 2 is that peaks in yield stress are observed in the broad temperature range of two-phase fields, and as ageing time increases, the peaks shift to the lower temperature of the $\alpha + DO_3$ two-phase field. Thus precipitation of α in the DO_3 matrix gives rise to hardening during ageing.

Microstructural changes with ageing were studied to confirm the presence of α precipitate. Fig. 3a and b show micrographs of specimens aged at 510 °C for 10 and 30 min, respectively. The broad dark line running through each figure is B2-type APB coated with a continuous layer of α phase, and the fine lines are DO_3-type APBs, coated with a thin layer of α in (a) and a thick layer of α in (b). It must be emphasized that fine α particles in DO_3 domains appear to a greater extent in the specimen aged for longer times.

Other examples are shown in Fig. 4a and b which are micrographs of the alloy aged at 540 °C for 0.1 and 100 min, respectively. The specimen aged for 0.1 min shows DO_3 domains without α precipitates, while the specimen aged for 100 min shows a high density of fine α precipitates in the DO_3 domain and a continuous layer of α on APBs. Precipitation of α either in a DO_3 domain or on APBs after ageing for a long time agrees well with the results reported by Allen [9] for 24% Al alloy. Therefore, hardening due to α precipitation must have an effect on the anomalous yield behaviour in the hypo-stoichiometric alloys.

However, it is noteworthy that in the case of 0.1 min ageing without α precipitates in DO_3 domains, an anomalous yield behaviour also appears in Fig. 2. Some careful research using 1 1 1 and 2 2 2 reflections

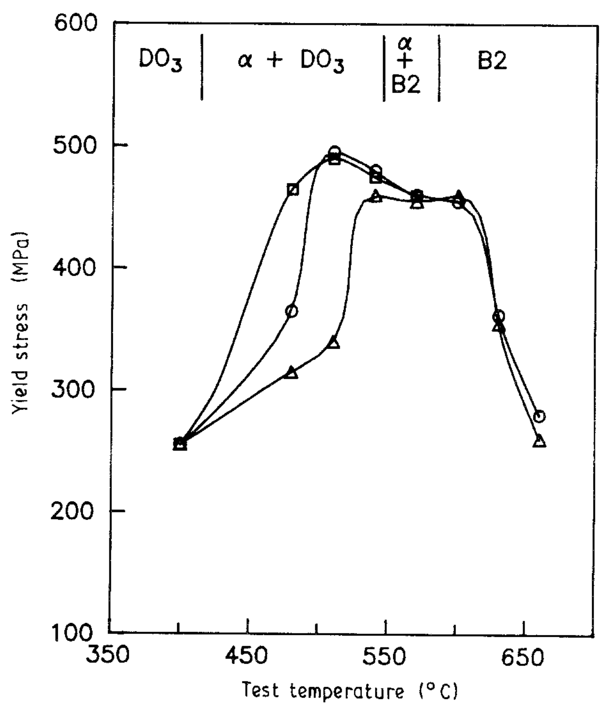


Figure 2 Temperature dependence of yield stress of 24.1% Al alloy. Ageing time (min): (△) 0.1, (○) 100, (□) 1000.

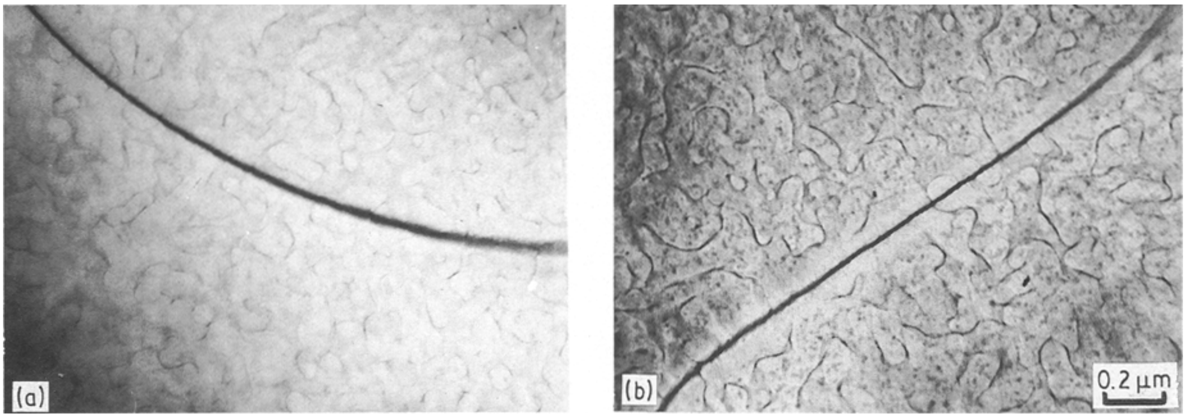


Figure 3 2 2 2 dark-field micrographs of 24.1% Al alloy, initially DO_3 ordered at 400 °C and aged at 510 °C for (a) 10 min, (b) 30 min.

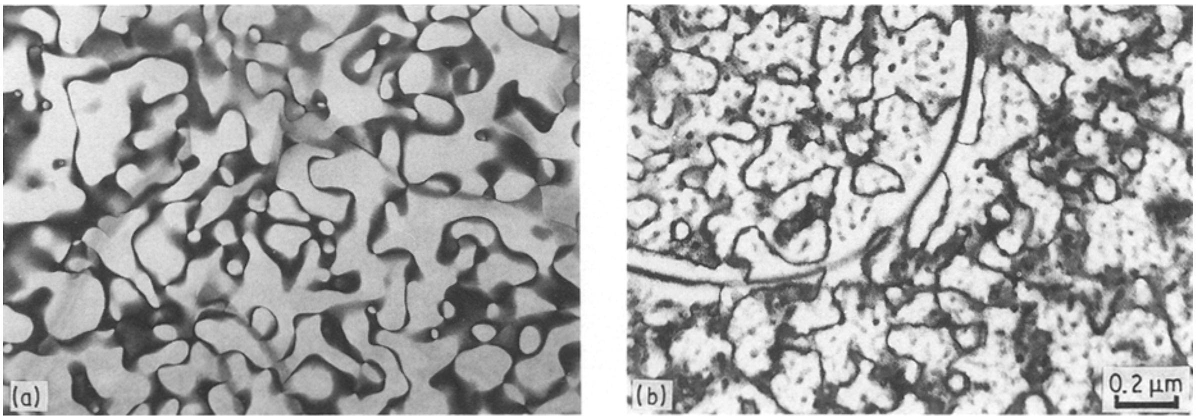


Figure 4 1 1 1 dark-field micrographs of 24.1% Al alloy, initially DO_3 ordered at 400 °C and aged at 540 °C for (a) 0.1 min, (b) 100 min.

revealed that a layer of α on the B2-type APBs existed in specimens aged for 0.1 min above 540 °C. However, it is believed that this has only a little effect on the anomalous yield behaviour because density of B2-type APB is very low, as shown in Figs 3 and 4, and a positive temperature dependence also appears at low temperature where distinct evidence of the α layer was not found in specimens aged for short times. Another feature of microstructural change during ageing was that the average diameter of the DO_3 antiphase domain increased with increasing temperature even in the case of 0.1 min, e.g. from 0.125 μm at 400 °C to 0.145 μm at 540 °C.

These results confirm that local redistribution of atoms in the DO_3 superlattice should occur during rapid heating and short term ageing. Therefore, it is believed that a decrease in the DO_3-type order, which can be achieved by local rearrangement of atoms through short-range diffusion, is responsible for the anomalous temperature dependence for the short term ageing. On the other hand, precipitation of α, which requires long-range diffusion, must be responsible for the additional behaviour such as the shift of the positive temperature dependence region and the peak temperature to the lower temperature side for the long ageing times.

The effects of ageing time on the anomalous yield behaviour of 27.2% Al alloys are shown in Fig. 5. The most important feature in this figure is that most of the anomalous behaviour is observed in the DO_3 region where neither α precipitation nor B2 formation can occur. Thus, the anomalous behaviour should not

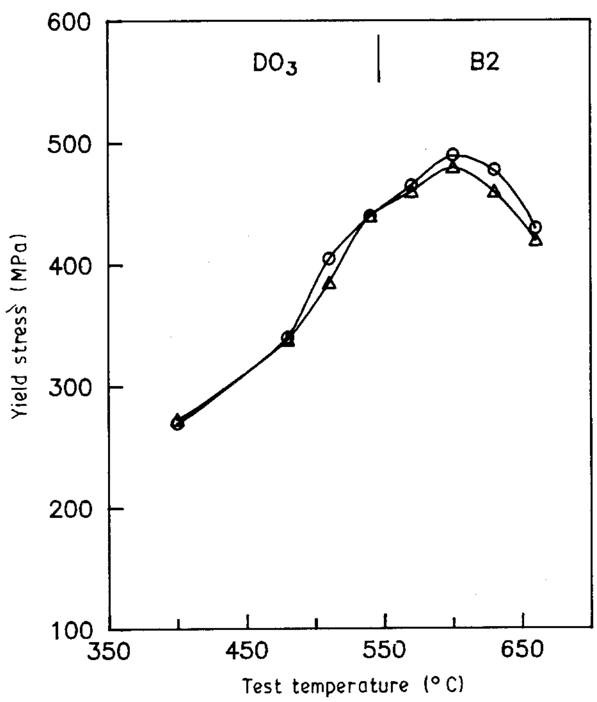
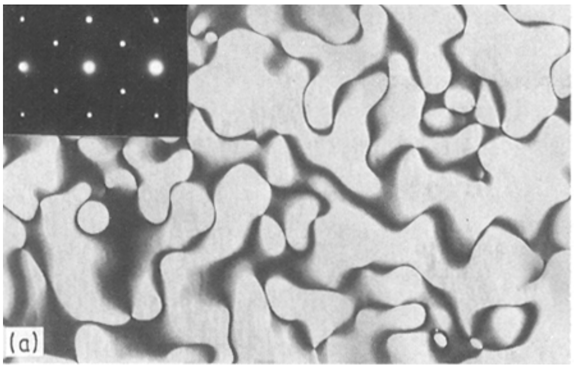
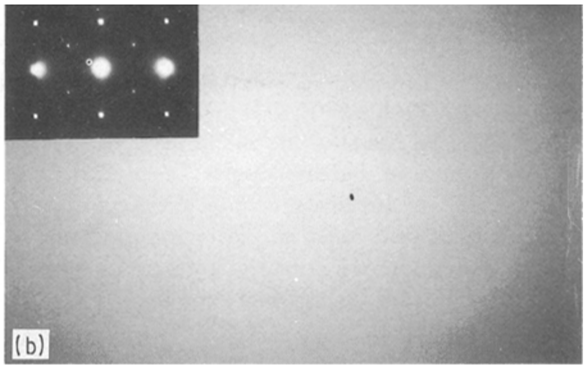


Figure 5 Temperature dependence of yield stress of 27.2% Al alloy. Ageing time (min): (△) 0.1, (○) 100.





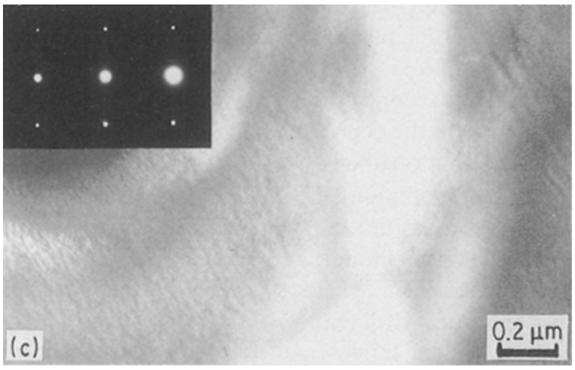


Figure 6 Dark-field micrographs of 27.2% Al alloy, (a) DO_3 ordered at 400 °C for 50 h, (b) subsequently aged at 600 °C for 0.1 min, both 1 1 1 DO_3 reflection, and (c) aged at 600 °C for 100 min, 1 1 1 B2 reflection.

be related to the α precipitation and B2 formation, i.e. Inouye's explanation [4] cannot be applied to this second-order transformation alloy.

Another important feature in Fig. 5 is that all the peaks are seen to be in the B2 region above 550 °C, different from the results for the hypo-stoichiometric alloy shown in Fig. 3. According to Lawley and Cahn [10] and Oki *et al.* [11, 12], the degree of DO_3-type order in Fe_3Al alloys begins to decrease near 400 °C, and becomes 0 at the DO_3–B2 transformation temperature. The degree of B2-type order also decreases to 0 in the higher temperature range above the transformation temperature. Therefore, it is considered that the anomalous behaviour in the DO_3 region is associated with the degree of DO_3-type order, and the behaviour in the B2 region is related to the decrease in the degree of B2-type order.

In Fig. 5, it should also be noted that the anomalous yield behaviour is nearly independent of ageing time, and so it exhibits similar trends in both cases. This behaviour in hyper-stoichiometric alloy is greatly different from that in hypostoichiometric alloy. Micrographs obtained from 27.2% Al alloy in order to investigate the variation of the microstructure with ageing are presented in Fig. 6. Well-developed DO_3 domains and intense 1 1 1 DO_3-type superlattice reflections are shown in Fig. 6a, a micrograph of a DO_3 ordered specimens at 400 °C. On the contrary, subsequent ageing in the B2 region results in the disappearance of DO_3 APBs not only on long-term ageing, but also on short-term ageing, as shown in Fig. 6b and c, which are micrographs for specimens aged at 600 °C for 0.1 and 100 min after DO_3 ordering, respectively. At the same time, the intensity of 1 1 1 DO_3-type superlattice reflections becomes weaker and weaker as ageing time increases, while the B2-type superlattice reflections remain almost constant.

Thus the decrease in the degree of order is so fast that the degree of DO_3-type order can decrease greatly even on short-term ageing. Accordingly, it is believed that the similarity in the anomalous yield behaviour between the short-term ageing and the long-term ageing comes from the rapid decrease in the degree of order during heating and ageing. These results also confirm that the previous result for the hypo-stoichiometric alloy aged for 0.1 min is associated with the decrease in the degree of order.

4. Discussion

The present study clearly shows that there are distinct differences in the anomalous yield behaviour between first- and second-order transformation alloys for Fe_3Al. In the first-order transformation alloy the anomalous behaviour is influenced by the changes in the degree of order before phase separation, and by the α precipitation after phase separation. In contrast, the behaviour in the second-order transformation alloy is caused only by the changes in the degree of order.

There are various explanations for the positive temperature dependence of yield stress in Fe_3Al alloys. The cross-slip mechanism presented originally by Kear and Wilsdorf [13] and extended to Fe_3Al by Hanada *et al.* [3] is a thermally activated process, so the anomalous behaviour due to cross-slip should occur with variations of thermal energy. However, anomalous behaviour can also appear at constant temperature only if the degree of order changes [14, 15]. In addition, differences in the behaviour between first- and second-order transformation alloys cannot be explained by the cross-slip mechanism.

Inouye's explanation [4] which is based on the precipitation of α cannot be applied to the anomalous behaviour in the second-order transformation alloy because there is no α precipitation. Moreover, because anomalous behaviour begins before B2 formation in both hypo- and hyper-stoichiometric alloy, his concept based on B2 formation is also unsatisfactory as an explanation for the whole anomalous behaviour in both compositions. According to Swann *et al.* [7], DO_3–B2 transformation occurs not by the classical nucleation–growth process but by a continuous process in which continuous changes in the degree of order take place homogeneously throughout the matrix. This transformation occurs only in a very narrow range of DO_3–B2 transformation temperature. Therefore, B2 formation hardly results in precipitation hardening, different from α precipitation. Accordingly, only the explanation based on α precipitation can be applied to a limited extent to the anomalous behaviour in the first-order transformation alloy after phase separation.

The APB generation model or dislocation transition model proposed by Stoloff and Davies [2] predicts that a peak in yield stress will appear just below the order–disorder transformation temperature (550 °C in their paper) which, in fact, they had observed. According to the present results, however, the peak appears primarily below 550 °C in the first-order transformation alloy, while it appears above 550 °C in the second-order transition alloy. It is considered that this contradiction stems from the difference in the phase transformation mode. The Stoloff–Davies theory is based on the homogeneous ordering process, but their alloy with 24.6% Al belongs to the first-order transformation alloys which are age-hardenable. Therefore, peaks in their experiment may be due, in part, to the precipitation hardening of α.

Nevertheless, the basic idea of the Stoloff–Davies theory can be applied to the explanation of the present results, because changes in the degree of order have a great influence on the anomalous yield behaviour not only in the second-order transition alloy but also in the first-order transition alloy before phase separation. Application of their theory, however, requires some modification because peaks in yield stress appear in the B2 region just above the DO_3–B2 transformation temperature.

Marcinkowski and Brown [16] showed that superdislocations in the DO_3 superlattice consist of four $a'/4\ \langle 1\,1\,1\rangle$-type ordinary dislocations bound together by the nearest neighbour antiphase boundary (NN APB) and next nearest neighbour antiphase boundary (NNN APB), and superdislocations in the B2 superlattice consist of a pair of $a/2\langle 1\,1\,1\rangle$ coupled by NN APB alone.

In highly ordered DO_3 structure, perfect dislocations are nucleated, and deformation can occur at low stress levels because there is no production of APB, whereas deformation in weakly ordered DO_3 structure can occur at higher stress levels due to trailing NNN APBs, which results in an increase in yield stress below the DO_3–B2 transformation temperature, as shown in Fig. 4. As the temperature increases above the DO_3–B2 transformation temperature, the B2-type degree of order decreases and yield stress increases due to the generation of imperfect B2-type superdislocations accompanied by the NN APBs. Further increase of temperature decreases APB energy and enhances diffusion-involved motion of dislocation, so that yield stress decreases after the peak in yield stress. The same explanation can be applied to the anomalous yield behaviour of the first-order transition alloy before phase separation.

5. Conclusions

1. Anomalous yield behaviour revealed great differences depending on the transformation modes.
2. In the first-order transformation alloy, precipitation of α phase had a great influence on the anomalous behaviour after phase separation, while changes in the degree of order played an important role before precipitation of α.
3. In the second-order transformation alloy, only the change in the degree of order was a principal factor in the anomalous behaviour.
4. The anomalous behaviour due to changes in the degree of order can be explained by a modified Stoloff–Davies model; transition from DO_3-type superdislocation to B2-type superdislocation and further to unit dislocation.

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