1-1-2014

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Abstract

We report an abnormally high residual dislocation density in aluminium in an Al/Ti/Al laminate annealed at 873 K for seven days. The residual dislocation density reaches 7.5 x 1014 m−2, higher than that in aluminum after severe plastic deformation processes such as accumulative roll bonding and high-pressure torsion. It is proposed that the high residual dislocation density may result from obstruction of the movement of TiAl3 nanoparticles by the grain boundary and Ti atoms conglomerating at vacancies distributed in the aluminium matrix at a high temperature for a sufficient time to allow a relatively stable crystal.

Keywords

dislocation, aluminium, void, composite laminate, annealing, plastic deformation

Disciplines

Engineering | Science and Technology Studies

Publication Details

Yu, H., Tieu, A. Kiet., Lu, C. & Kong, C. (2014). Abnormally high residual dislocation density in pure aluminum after Al/Ti/Al laminate annealing for seven days. Philosophical Magazine Letters: structure and properties of condensed matter, 94 (11), 732-740.

Abnormally high residual dislocation density in pure aluminum after Al/Ti/Al laminate annealing for seven days

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We report an abnormally high residual dislocation density in aluminium in an Al/Ti/Al laminate annealed at 873 K for seven days. The residual dislocation density reaches 7.5×10^{14} m^{-2}, higher than that in aluminum after severe plastic deformation processes such as accumulative roll bonding and high pressure torsion. It is proposed that the high residual dislocation density may result from obstruction of the movement of $TiAl_3$ nanoparticles by the grain boundary and Ti atoms conglomerating at vacancies distributed in aluminium matrix at high temperature for a sufficient time to allow a relatively stable crystal.

Keywords: dislocation; void; composite laminate; annealing

1. Introduction

Dislocation, a classic topic in material science because of its significance in determining the mechanical properties of materials, has received considerable attention from researchers. Dislocation density is a critical material variable that determines dislocation mobility, strength and ductility of materials.

In the absence of recovery, the dislocation density in metals increases with an increase in strain, as shown in Eq. (1): **[1]**

$$\rho_g = (bl)^{-1}\varepsilon \tag{1}$$

where ρ_g is the total generated dislocation density, b the Burgers vector, l the mean free path for the dislocation movement, and ε the strain. Based on the theory, some Severe Plastic Deformation (SPD) techniques have been developed to produce

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nanostructured metals **[2-5]**. After an SPD processing, the residual dislocation density in metals is generally higher than that before processing **[6,7]**, however, further increase in the strain can lead to grain coarsening and dislocation annihilation **[8]**. Because the dislocation multiplication resulting from straining is balanced by dislocation annihilation due to grain boundary migration, strain-induced structural evolution ceases, and the dislocation density reaches its limit, which in heavily deformed metals is typically ~10^{15} m^{-2}.

The dislocations existing in crystalline solids are thermodynamically unstable entities. In general, low temperature could lead to a higher density of dislocations to effectively suppress the dynamic recovery **[9, 10]**. Yu et al **[11]** found that the residual dislocation density of aluminum after freezing (to about 253 K) is 7.5×10^{13} m^{-2}, and at room temperature it is 5.6×10^{13} m^{-2}. The dislocation density can be reduced by annealing **[12]**. Deformed metals are annealed to rearrange dislocations and relieve stresses. Long-duration annealing at high temperatures, which promotes the approach to thermodynamic equilibrium, results in a diminution of the dislocation density. During isothermal annealing, the dislocation density ρ varies with time t according to Eq. (2): **[13]**

$$1/\rho = 1/\rho_0 + At \tag{2}$$

where ρ_0 is the initial dislocation density and A is a constant. Ma et al **[14]** found that the dislocation density in the grain interior decreased by 60% after annealing. Zhang et al **[15]** found that the residual dislocation density in an HPT-processed pure aluminum sample after annealing was about 7×10^{13} m^{-2}. Mizuno et al **[16]** found that the residual dislocation density of the specimens decreased to 1×10^{7} m^{-2} in ultrahigh-purity aluminum single crystals by cyclic annealing.

In recent years, composite laminates have attracted significant attention **[17-19]**. During heat treatment, some metallic laminates such as Ti/Al and Cu/Ti, are transformed into metal-intermetallic laminated composites. These offer an attractive combination of properties such as high toughness, low density, and high oxidation resistance of the intermetallics. Metal-intermetallic laminates can perform various

desirable functions such as ballistic protection, heat exchange, blast mitigation and vibration damping **[20]**. Compared with pure metals, the residual dislocation density in composite materials is much greater. At high magnification, the residual dislocation density of the matrix was observed to range from 1×10^{14} to 4×10^{14} m^{-2} by Arsenault and Fisher **[21]**, who pointed out that the dislocations are presumably generated by stresses resulting from differential contraction between SiC and the AA6061 aluminum matrix. On cooling to about 500 K, dislocations reappeared at the interfaces of large particles, forming tangles in the vicinity of the precipitate. Vogelsang et al **[22]** found that the microstructure of the 20% whisker sample before heating is characterized by small subgrain size (2 to 3 μm) and by a residual dislocation density of about 10^{13} m^{-2}.

The intermetallic compounds of the Ti-Al system and alloys based on such compounds have great potential for applications in the aerospace and automotive industries because of their light weight and excellent high-temperature properties, advanced creep characteristics, and good corrosion resistance. They have long been identified as next-generation structural materials and investigated extensively.

In this letter, we report the fabrication of Al/Ti/Al laminate foils by asymmetric rolling, followed by annealing at 873K for up to seven days. We found an abnormally high residual dislocation density in the pure aluminum zone. The residual dislocation density was found to reach 7.5×10^{14} m^{-2}, higher than that in aluminum after being subjected to severe plastic deformation processes.

2. Experimental Investigation

300 μm thick pure aluminum AA1235 and 25 μm thick pure Ti foils were used in the experiments. These were cold-rolled using a four-high rolling mill with 50 mm diameter work rolls. The foils were stacked as Al/Ti/Al and welded at one end to prevent slippage. The composite 625 μm thickness laminates were rolled to reduce the thickness to 130 μm. After rolling, the laminate foils were annealed at 873 K for 6 h, 12 h, 48 h and 168 h (7 days), and left to cool naturally in air. The Al/Ti interfaces in the rolled and annealed laminate foils were investigated. The RD-ND plane of the

rolled samples was used to obtain Transmission Electron Microscope (TEM) images. An FEI xT Nova Nanolab 200 Dual-beam workstation was used to prepare thin-foil specimens for further TEM observation. This provides a means of making a thin slice of 100 nm in a "site-specific" way. The thickness of the specimen is uniform, and the unevenness is less than 5%, as shown in Figure 1a. The specimens were placed on a standard carbon film Cu grid using an ex-situ lift-out method. A Philips CM200 Field Emission Gun Transmission Electron Microscope (FEG/TEM) equipped with a Bruker Energy Dispersive X-ray (EDAX) Spectroscopy system operating at an accelerating voltage of 200 kV was used to investigate the microstructure details. After the completion of all TEM imaging, we used a highly efficient Slicon Drift Detector (SDD) EDS detector combined with a weak electron beam (condenser aperture II No2) and very spot size (spot 7) for a short time (20 seconds for each point analysis, and 1.5 minute duration for the mapping). Under these conditions, it is unlikely that the high residual dislocation density is affected by the EDS process. The dislocation density is the cumulative dislocation length per unit volume. The direct measurement of the dislocation density was based upon bright field TEM micrographs of the region of interest. In this study, all measurements were done under two-beam conditions of {200} reflections near <001> zone axes. Some dislocations were invisible in the TEM images in certain orientations. So the measured dislocation density assumed homogeneous distribution of the tangled dislocations in 3D space **[23]**. The measurements were done in the region of 500 nm×500 nm×100 nm, and the reported value was the average of three measurements.

3. Results and discussion

Figure 1 shows an FIB sample and TEM micrographs at the frontiers of the $TiAl_3$ nanoparticles region near the Al layer under a range of annealing times. Upon annealing, the Ti and Al diffuse at the interfaces, and some of Ti and Al react to form intermetallics. Initially, a number of voids were observed in the annealed structures. These were caused by the Kirkendall effect and by the difference in molar volumes between the reactants **[24]**, as shown in Figure 1b. As the annealing time increased,

the voids at the $TiAl_3$/Al interface decrease, (Figure 1c), leaving a number of smaller voids at the $TiAl_3$ particle frontier. After annealing for 48 h, voids at the frontier of $TiAl_3$ are hardly noticeable (Figure 1d). However, after 168 h of annealing, we found an abnormally high residual dislocation density in the Al zone approaching the $TiAl_3$ particle, (Figure 1e). Figure 1f shows a detail of Figure 1e. In the Al zone approaching the $TiAl_3$, the residual dislocation density is about 7.5×10^{14} m^{-2}. The low-density region should have a residual dislocation density of 3.6×10^{13} m^{-2}, and the "dislocation-free" region a residual dislocation density up to 1.8×10^{12} m^{-2}. Generally, dislocations could be piled up neatly at the frontiers of an particle. However, a random distributed mass of dislocations within the whole grain was observed, as shown in Figure 1e. Ti and Al alloys can be synthesized as different intermetallics such as Ti_3Al, TiAl, $TiAl_2$, and $TiAl_3$ when processed at different temperatures. Generally, when the annealing temperature is less than 923 K, the titanium aluminide forms $TiAl_3$ at fairly low temperatures in the prior Al-rich layer after just a few hours **[25]**. The element distribution in a local zone of Figure 1e is shown in **Figure 2**. In Figures 2a, 2b, 2c and 2d, the intermetallic particle is seen surrounded by pure aluminium. In Figure 2e, the particle is $TiAl_3$. In Figure 2f, it is seen that the high dislocation density zone is aluminium with about 0.5% Ti. The particle size of the Region Of Interest (ROI) is about 500 nm. Referring to the profiles of previous EDS line scan study in similar working conditions, the distance of affected region due to the X-ray generated from the neighbouring grain is less than 100 nm for a vertical grain boundary. So we believe that a few Ti atoms did exist in the ROI.

Figure 3 shows the residual dislocation density in aluminium subjected to different processes. For pure aluminium, the residual dislocation density is very low after annealing. As the proportion of other elements increases, the residual dislocation density increases gradually. For an Al single crystal, the residual dislocation density is only 1×10^{7} m^{-2} **[19]**. With 20% whisker in pure Al, the residual dislocation density increases to 10^{13} m^{-2} **[22]**, and 4×10^{14} m^{-2} for AA6061 with SiC particles **[21]**. Vogelsang et al **[22]** believed that the dislocation generation mechanism that could account for this high dislocation density should be based on the large difference in the

thermal expansion coefficients, which might result in high stress at the boundary particle/matrix. For Al and SiC, the thermal expansion ratio is 10:1 **[21]**. When the composite is cooled from elevated temperatures, uneven strains occur due to differential thermal contraction at the Al/SiC interface, which can generate dislocations. Table 1 lists the thermal expansion coefficients of Al and $TiAl_3$ from 300 K to 900 K **[26, 27]**. In this temperature range, the ratio of thermal expansion coefficients between Al and $TiAl_3$ varies from 0.95 and 1.2. In addition, the residual dislocation density in pure Al has been found to increase greatly under SPD. The residual dislocation density of highly strained aluminium can reach 3.46×10^{14} m^{-2} and 7.07×10^{14} m^{-2} after HPT [6] and ECAP [6] processing respectively. Miyajima et al **[7]** found that the residual dislocation density was around 10^{14} m^{-2} in commercial pure aluminum samples when the equivalent strain is about 10 in samples subjected to ARB. However, all these values are less than the value found in the current study, which is 7.5×10^{14} m^{-2}.

In the current study, Al/Ti/Al laminates underwent severe plastic deformation during rolling, causing dislocations that were annihilated during annealing at the first stage. Figure 1b shows low residual dislocation density and some voids in the pure aluminium zone. The above discussion suggests that the high residual dislocation density in the pure alumnium zone is caused neither by the difference in thermal expansion coefficients nor by severe plastic deformation, but by a new mechanism.

It is well known that the annilation of the dislocations may result in vacancies and voids. Seitz **[28]** reported the mechanism of vacancy generation by moving dislocations. He proposed four mechanisms by which vacant lattice sites could conceivably be generated during plastic flow: 1) by local heating of the lattice in the immediate vicinity of the dislocation as it moves through the lattice and absorbs energy from the applied stress field; 2) by geometrical means in which dislocations of opposite sign moving in neighbouring planes annihilate one another and produce a row of vacancies; 3) as a result of instability of the in-phase motion of atoms during passage of a dislocation; 4) as a result of large, transient thermal pulses in regions where dislocations annihilate one another and produce a large lattice disturbance. The

theory of vacancies and voids caused by the movement of dislocations is widely used in fracture analysis **[29]**. However, there has never been any report on the vacancies and voids transferring into dislocations during annealing.

In the present study, we propose that the high residual dislocation density in the aluminium zone is related to the evolution of voids and the diffusion of Ti atoms. The diffusion between Ti and Al and formation of intermetallic compounds during the long-time annealing of severely deformed material makes it possible for some Ti atoms to move beyond the boundary of the stoichiometric $TiAl_3$ region and form a buffer zone of a low-concentration solid solution of Ti in Al at the frontier region of $TiAl_3$ particle, as shown in Figure 2f. **Figure 4** illustrates the generation of high dislocation density in the aluminium matrix. Voids are caused by the difference in speeds at which the Al and Ti atoms move in the initial stages of diffusion (Figure 4a). As the annealing time increases, large Kirkendall voids are divided into smaller voids and distributed in the Aluminium matrix (Figure 4b). For metallic materials, the number and size of voids could be reduced by increasing the time of exposure to high temperature **[30]**. According to Fick's second law of diffusion, with an increase in time, there is sufficient opportunity for atomic diffusion to occur around the voids. Pines and Syrenko **[13]** believed that when the microcracks are healed, dislocations begin to act as vacancy sources. This results in an acceleration of their climb and consequential annihilation. As the annealing time increases, irregularly shape voids will gradually change into spherical voids to minimize the surface energy **[31]**. In addition, some of the Ti atoms will move and be adsorbed at the void surface (Figure 4c). When $TiAl_3$ particles meet a grain boundary, this will stop further movement of the $TiAl_3$ particles. Only a few Ti atoms pass through the grain boundary. With the movement of atoms at high temperature, the size of the spherical voids will gradually decrease and become vacancies **[32]**. In addition, the distributed Ti atoms will assemble at the vacancies to minimize the free energy (Figure 4d). Vacancies have a tendency to cluster at the boundary. As clusters increase in size, they eventually change to other configurations which can be interpreted as nucleation of dislocation loops **[33]**. In addition, Wei et al **[34]** found that the dislocations generated during a

5.957 nm-length crack healing through a molecular dynamics model. At such conditions, the sheets was cooled from 873 K to room temperature, and dislocations are generated and pinned by the Ti atoms or clusters yielding a dislocation density of 7.5×10^{14} m^{-2} (Figure 1e).

4. Conlcusions

When the annealing temperature of Al/Ti laminates is less than 873 K, only $TiAl_3$ intermetallics are developed when the annealing time is 168 h. The microstructure at the intermetallic $TiAl_3$ nanoparticle frontier changes greatly with annealing time. When the annealing time is 6 h, some large Kirkendall voids form at the $TiAl_3$ frontier. When the annealing time reaches 48 h, there are only some small distributed voids. As the annealing time increases to 168 h, an abnormally high residual density of dislocations (7.5×10^{14} m^{-2}) is observed.

Acknowledgements

The authors gratefully acknowledge the financial support from the Vice-Chancellor's Fellowship Grant and URC small grant at the University of Wollongong, and from the National Natural Science Foundation of China through Grant 51105071.

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Captions of Figures and a Table

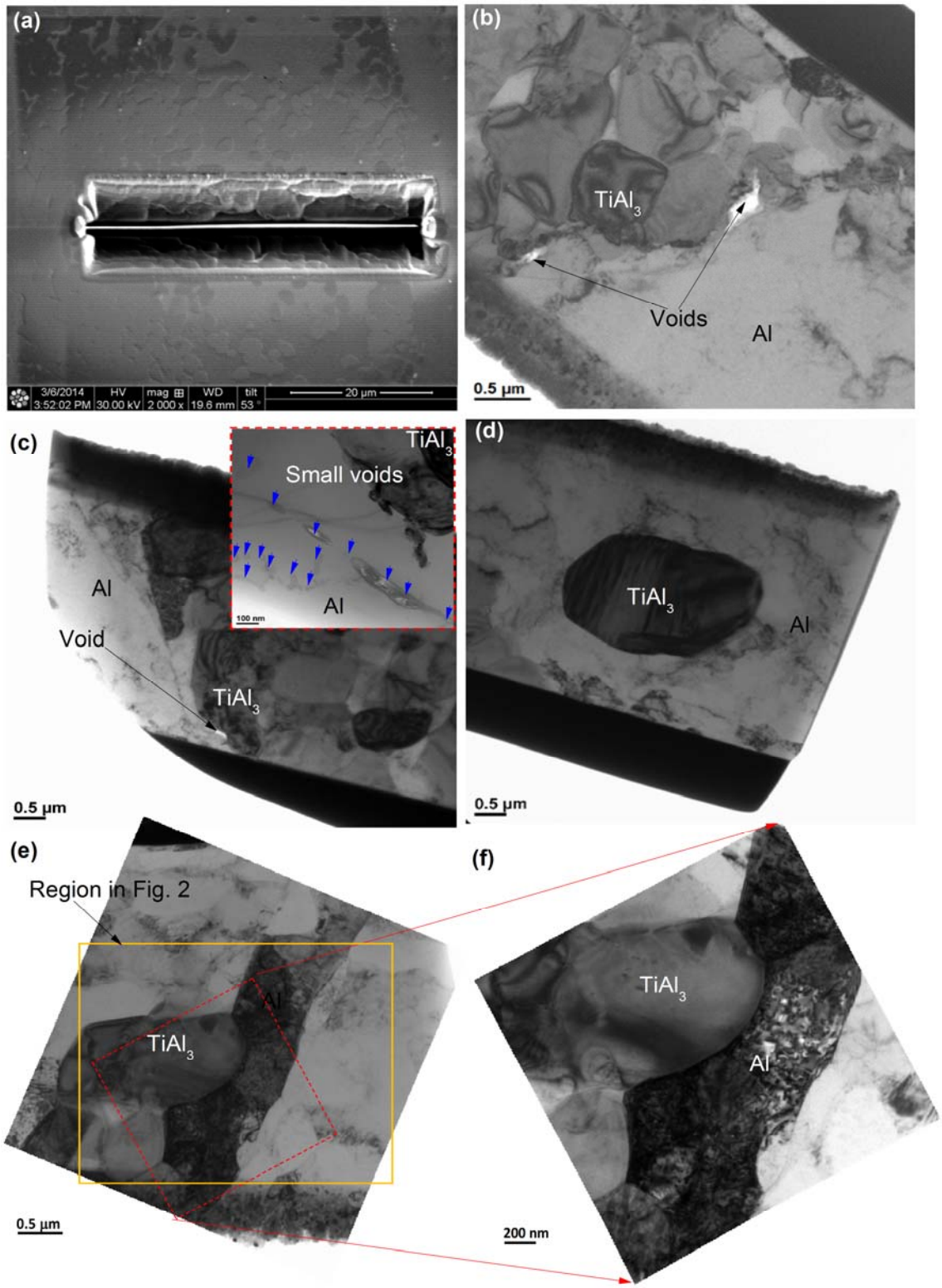

Figure 1. FIB image of Al/Ti/Al laminate for annealed 168 h (a); TEM images of annealed Al/Ti/Al laminates in $TiAl_3$ nanoparticle frontier for 6 h (b), 12 (c) 48 h (d) and 168 h (e); (f) is a detail of (e).

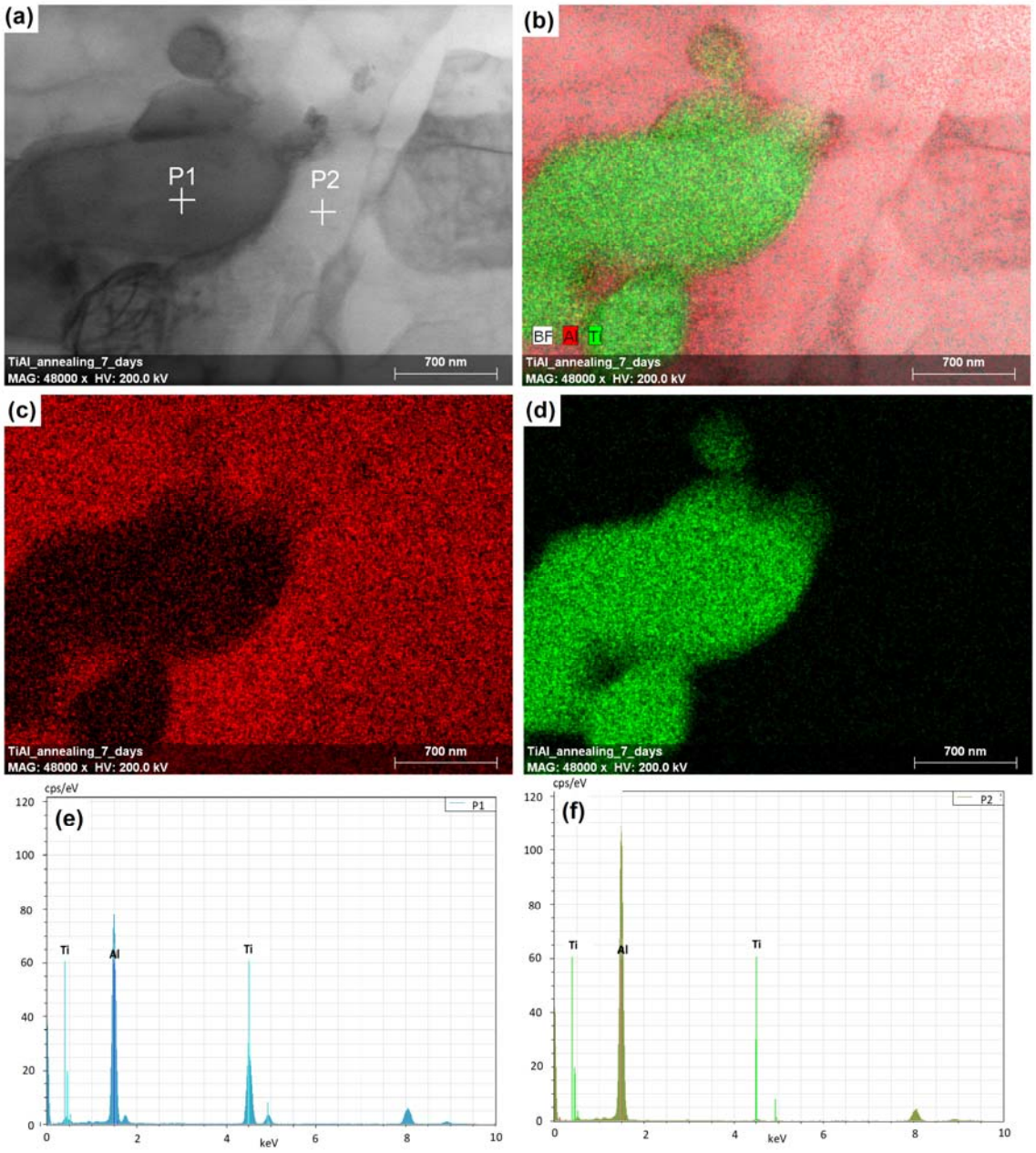

Figure 2. Element distribution at the local zone in Figure 1e. (a) Image of local zone in $TiAl_3$ nanoparticle frontier, (b) Combined Ti and Al distribution, (c) Al element distribution, (d) Ti element distribution, (e) element composition at P1 in Figure 2a, and (f) element compostion at P2 in Figure 2a.

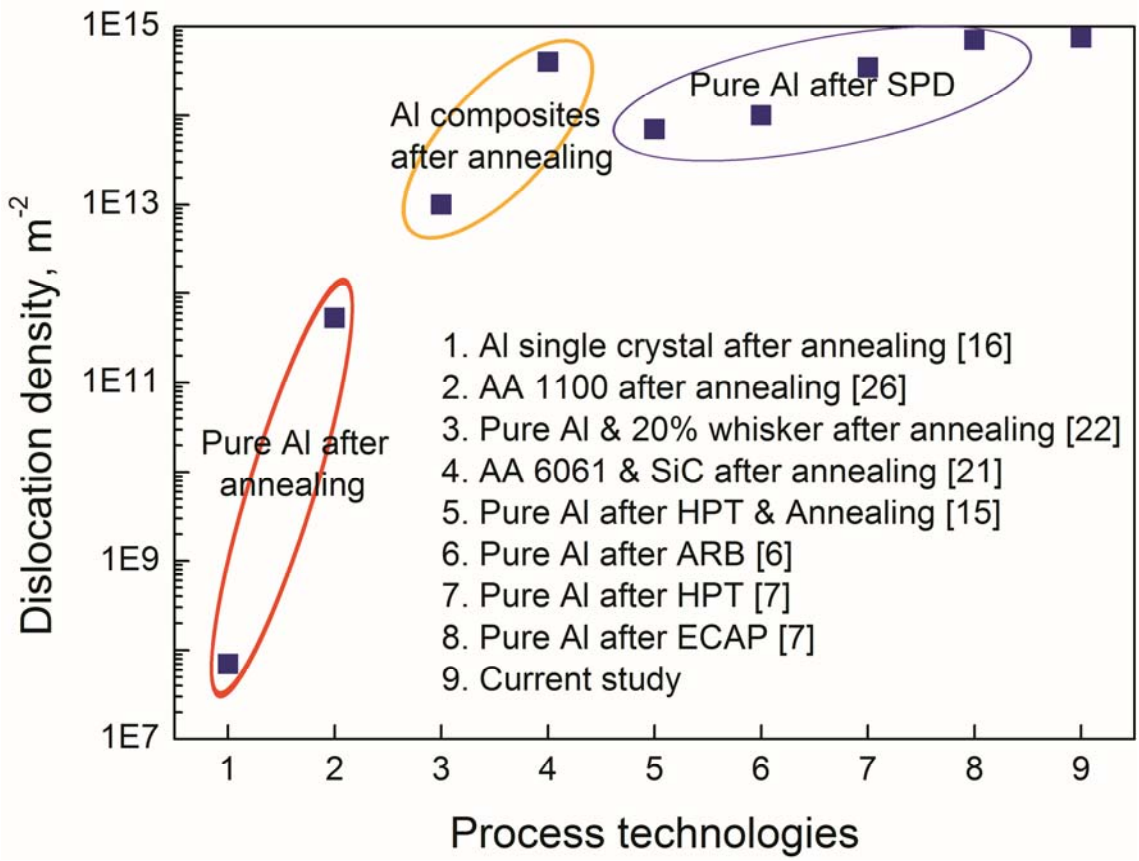

Figure 3. Dislocation density of Al after different process technologies.

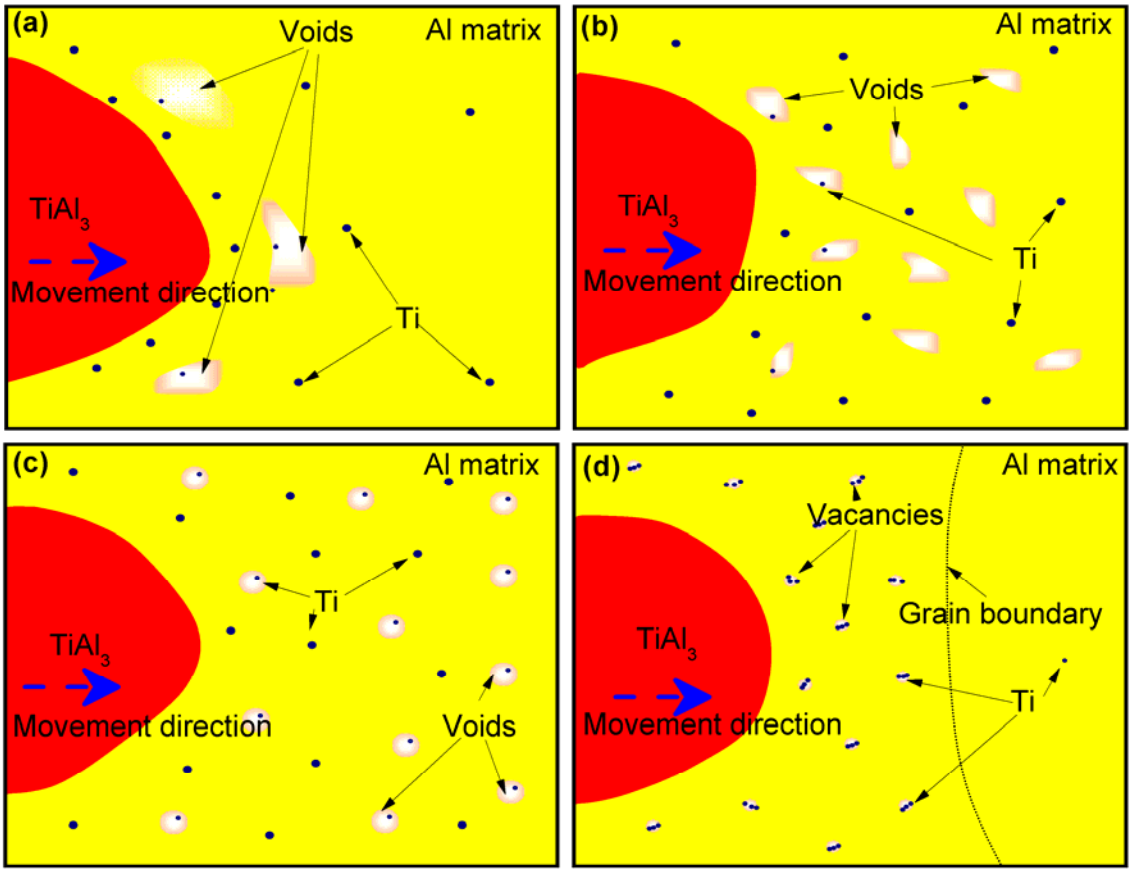

Figure 4. Schematic illustration of dislocation generation prcessing in aluminium under annealing of Al/Ti/Al laminates. (a) large Kirkendall voids at fronter of $TiAl_3$; (b) large voids divided into smaller voids; (c) voids with irregular shape changes into circular voids, and Ti element adsorbed into the voids; (d) voids change into vacancies, most of Ti elements adsorbed at the vacancies surfaces, and generate the high density of dislocations.

TABLE 1. Thermal expansion coefficient of Al and $TiAl_3$ at different temperatures.

Temperature, K	300	450	600	750	900
Al, 10^{-6} /K	23.2	24.5	25.1	25.2	25.6
$TiAl_3$, 10^{-6} /K	19.5	22.5	24.3	25.8	26.9