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Diffusion Bonding of Superplastic Aluminum Alloys

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Introduction

Ability to diffusion bond aluminum alloys, in particular superplastic aluminum alloys, will complete the technology-base that is strongly needed to enhance the use of superplastic forming (SPF) technology. Concurrent diffusion bonding (DB)-SPF is considered to be an energy-saving manufacturing process since it simplifies the production of complex components. Moreover, because of increased design flexibility, overall manufacturing cost and component weight are signficantly reduced.[1]

Diffusion bonding is an attractive manufacturing option for applications where the preservation of the base metal microstructure and, in turn, mechanical properties is imperative in the bond area. The process utilizes either the solid state or transient liquid phase (TLP) bonding to produce a bond with microstructure continuity in the joint.[2,3] In addition, there is no localized thermal gradient present to induce distortion or to create residual stresses in the component, thereby increasing structural integrity.

Despite the strong interest by both the aerospace [4] and automotive [5] industries to extend this technology to aluminum (Al) alloys, technical progress in this specific area has been relatively slow. So far, only certain titanium alloys have been demonstrated to be DB-SPF.[1] The extreme difficulty associated with the DB of Al alloys is a readily formed, tenacious Al oxide layer. Unlike Ti alloys, the solubility of oxygen in aluminum is less than 0.01 at-% and the stability of oxide is such that it will not decompose at desired bonding temperatures, even in vacuum. The oxide layer acts as an effective barrier preventing atoms from interdiffusing across the bond interface and, consequently, yields a poor metallurgical bond. However, the aluminum-lithium alloy 8090 has exhibited successful solid state diffusion bonding by modifying the Al oxide to less stable, discontinuous lithium-rich Li-Al spinels.[6] They appear as discrete, brittle particles on the bond interface. Joints that were solid state diffusion bonded in vacuum showed a planer microstructure at the interface and shear strengths similar to those of the base material. Moreover, the 8090 alloy has lower density, and higher specific strengths and modulus than the other high strength Al alloys, which are attractive attributes for weight-restricted aerospace applications. It is also quench-insensitive, which reduces the possibility of distortion to fabricated DB-SPF components,[7] and can precipitate strengthening phases during cooling.

Another Al alloy that has been studied for DB is 7475, which has high strength and good corrosion resistance.[8,9] In addition, it can be thermomechanically processed to be superplastic. However, the DB successes for both 7475 and 8090 have been limited to the laboratory conditions only. As the technology moves from the laboratory to production, the DB process has to be production feasible and cost-effective.

A comparative study of the DB of SPF Al alloys has been initiated at the Lawrence Livermore National Laboratory to determine the effect of surface chemistry on the DB properties of the aluminum alloys, 8090 (2.4Li-1.18Cu-0.57Mg-0.14Zr-Al) and 7475 (1.26Cu-2.01Mg-5.27Zn-0.17Cr-0.006Si-0.012Fe-Al). The integrity of the diffusion bonds was evaluated for both interlayered and bare surfaces. Two interlayer elements, copper (Cu) and zinc (Zn), were compared. Although the eutectic temperature of Al-Cu is 548°C, a thin Cu layer in contact with 8090 has been shown to lower its eutectic temperature to ~521°C.[6] In 8090, Cu is one of the primary alloying elements but has a limited solubility in Al at the bonding temperature. Zinc, on the other hand, forms a considerably lower eutectic (380°C) with Al and is highly soluble in Al. In 7475, Zn is the primary alloying element. The diffusivity of Zn in Al is much faster than that of

Cu, but Zn forms a more thermodynamically stable oxide. These subtle metallurgical differences will affect the TLP formation at the interface, which in turn will influence the bond quality.

Experimental Procedure

The as-received plates of 8090-T81 and 7475-T6 were 9.5 mm and 19.0 mm thick, respectively, and were cut to 25.4 mm x 25.4 mm samples. Plates were used to determine the bond tensile strength. The surfaces to be bonded were machined and lapped with 600 grit SiC paper. To enhance grain boundary diffusion at the bonding interface, the bonding surfaces were matched in the short-transverse orientation to take advantage of elongated grains. Figure 1 shows the microstructure of 8090 and 7475 in three orientations.

Electroplating process was used to coat Cu and Zn interlayers on the Al surfaces. Prior to plating, the samples were degreased in acetone and immersed in Enthone NS-35 solution for 5 min. Then 5 sides were masked. The unmasked side of the samples was again electrocleaned in Enthone solution, followed by immersing in a 50 vol.-% nitric acid for 10 min. To further remove any residual oxide film on the surface, the samples were again immersed in a 30 vol.-% zincate solution containing ZnO/NaOH for 2 min. A 10 μm thick layer of Cu was plated on the bonding surfaces using an Allied Kelite solution at 2.3 A/m^2 (25 A/ft^2), while a 10 μm thick layer of Zn was plated using a Zn cyanide bath at 1.4 A/m^2 (15 A/ft^2), followed by water rinse.

The diffusion bonding apparatus consisted of two ceramic platens each 50 mm diameter and 150 mm thick, a quartz tube 63.5 mm diameter, weights, and a water-cooled, tubular quartz furnace. Before putting the surfaces together, the samples were again ultrasonically cleaned in ethanol and then, they were sandwiched between ceramic platens prior to placing inside the quartz tube. In the furnace, the tube was sealed and back-filled with argon and a predetermined weight, which was held in place by the movable cross-head of a test machine, was loaded on the ceramic platens. Two chromel-alumel thermocouples were used to monitor the temperature, one embedded in the sample and other near the sample for furnace temperature control.

The thermal history used for the DB of both 8090 and 7475 samples was the same. Under a static pressure, the samples were heated to 550°C at 18°C/min and held for 15 min to ensure melting of Cu interlayer, lowered to 500°C for 60 min, and then slowly furnace-cooled. For the 7475 samples, a higher applied pressure was used, 2.9 MPa (420 psi), in comparison to the 8090 samples, 1.2 MPa (175 psi). The bonded interfaces and fracture surfaces were examined using optical metallography and scanning electron microscopy (SEM) fractogrphy,, respectively. The bond quality was determined by tensile tests at room temperature. The specimens were pulled at a constant crosshead speed of 1.27 mm/min with the bond located at the center of the 25.4 mm gage length.

Results and Discussion

Diffusion Bonding of 8090

For the bare surface DB of 8090, the benefit of Li-Al spinel formation at the bonding surfaces was not found. In a non-vacuum environment, the specimen simply did not bond. Although there was a sufficient pressure available to produce interfacial contact at the bonding surfaces, in an argon atmosphere with a partial pressure of oxygen, even the freshly exposed metal surfaces would reoxide and prevent the metal/metal contact required to form the bond.[10] Figure 2 shows the heavily oxidized fracture surface of DB 8090 with localized dimples suggesting an earlier presence of spinel particles.

The use of thin protective layer to prevent oxidation on the joining surfaces has been utilized in many applications in various industries. For example, in the electronic industry, maintaining the solderability of Cu lands on printed circuit boards is critical during component packaging.[11] To protect these Cu lands, a thin layer of a eutectic lead-tin solder is commonly coated on the Cu. During packaging, the solder dissolves into the bath, providing oxide-free Cu surfaces to be resoldered. A similar role applies to the interlayer, which is to protect the surfaces prior to bonding. At the bonding temperature, melting at the interlayer interface forms a TLP. However, the desorption, transportation, and reprecipitation phenomenon that occurs at the melt region [12] will vary depending on the reaction between substrate and interlayer.

The difference in the Cu and Zn TLP formation and their effects on DB are shown in the cross-sections of the polished DB joints of 8090 (Figure 3). For the Cu interlayer, the bond interface was difficult to detect (Figure 3a). A discrete dispersion of Cu-rich phases near the bond area provided the only observable indication of the bond location. Three ternary intermetallic compounds that are in equilibrium with the Al-rich solid solution are T_B ($Al_{7.5}Cu_4Li$), T_1 (Al_2CuLi), and T_2 (Al_2CuLi_3).[13] These particles were only present within 125 μm from the interface. A similar observation was made by Bushby and Scott [14] where they found that Cu had diffused up to 150 μm into the Al when held at 550°C for 100 min. Also note that these Cu-rich phases did not form on all the grain boundaries. Diffusion of Cu or TLP into the base metal through the grain boundary region is strongly influenced by the grain misorientation, whether it is random (high-angle) or ordered (low-angle) boundaries.[15] In the case of the random high-angle boundaries, higher grain boundary energy promotes deeper liquid penetration and the formation of a smaller dihedral angle with the liquid phase. On the other hand, a much smaller effect of liquid penetration and change in the solid-liquid interface profile is seen with low-angle boundaries based on their lower boundary energy.

For the Zn interlayer specimens, the reaction at the interface was significantly different. The cross-section of the DB specimen showed more distinct outline of the bond interface (Figure 3b). Unlike Cu, Zn does not form a common ternary intermetallic compound with any of the alloying elements in 8090, but in contact with Al, undergoes a phase change at ~ 385°C,[2] which is close to the eutectic temperature of the Zn-Al system. As the Zn-rich TLP dissolves rapidly into the Al matrix at the DB temperature, the areas with eutectic composition at the interface will be the last to solidify. Thus, the noticeable difference in composition was only seen at the bond interface of an unetched specimen. Both Cu and Zn interlayer specimens contained nonplanar interface profiles and the presence of extensive plastic deformation was evident in the grains near the interface where they were severely misaligned. Thus, the DB pressure of 1.2 MPa used for 8090 was sufficient for intimate contact to occur at the interface.

In order to effectively compare the bond quality, the 8090 base metal was reprocessed using the DB parameters. The tensile test results are listed in Table 1. With the same interlayer thickness and bonding parameters, the tensile properties of the Cu interlayer specimens of both alloys were found to be substantially better than those of the Zn interlayer specimens. For the Cu interlayer 8090 specimens a joint efficiency, which is a ratio of the joint yield strength to the base metal yield strength, of 50% (200 MPa) was obtained. In comparison, only a 25% joint efficiency was obtained for the Zn interlayer 8090 specimens.

Figure 4 shows the fracture surfaces of the Cu and Zn interlayer specimens. The typical fracture behavior of 8090 alloy was seen in the fracture surface of a Cu interlayer specimen. The surface revealed ductile dimples with delamination along the grain boundaries (Figure 4a). Because the delamination occurred along the boundaries that contained Cu-rich phases, the grains shown in the fracture surface appeared much larger than those in the Zn interlayer fracture surface (Figure 4b). In the case of the Zn interlayer specimens, the crack propagated along the bond interface resulting in predominately intergranular fracture with some ductile dimples. As a result, this fracture surface is more representative of the pancake-like microstructure of the base metal.

Diffusion Bonding of 7475

For the 7475 alloy, the Cu interlayer specimens displayed promising bond strengths of 175 MPa. The cross-section of the Cu interlayer specimen showed a high fraction of metal-to-metal contacts, where the bond interface was not easily discerned, as indicated by the arrows in Figure 5a. However, there were enough areas with inadequate bonding to adversely influence the bond strength. In addition, a slow cooling of the specimen from the DB process resulted in the precipitation of fine $MgZn_2$ in the matrix and Al_2CuMg, or Al_7Cu_2Fe, at the grain boundaries.[16]

The precipitation of grain boundary phases led to a solute-depleted, precipitate-free zone (PFZ) for the grains near the bond interface. A similar PFZ developed during SPF when diffusional creep was the dominant deformation mechanism.[17]

Diffusion bonding of 7475 with the Zn interlayer continued to have inadequate bond strengths where many of the specimens broke during handling. Thus, higher applied pressure of 2.9 MPa for the DB 7475 had no positive effect on the bond strengths. Instead, the bonding temperatures of 500 and 550°C used for the DB specimens with the Zn interlayer may have been too high, since Zn undergoes a phase change at 385°C in Al [2] and is more susceptible to oxidation than the Cu. In addition, as the Zn prematurely diffuses into the matrix, the areas that have not established an interfacial contact is exposed to an argon atmosphere with a partial pressure of oxygen. Many of the fracture surfaces were heavily oxidized.

The fracture behavior of the DB 7475 specimens with the Cu interlayer was significantly different from those of the DB 8090 specimens (Figure 5b). Fracture occurred predominantly intergranular in the base metal with some decohesion between the substrate and previous TLP. The grain surfaces were covered with both deep and shallow dimples. A qualitative analysis using energy dispersive spectroscopy of the fine particles in the boundaries indicated that they were rich in Cr. Chromium is purposely added to Al to form $CrAl_7$ dispersoids which were to function as the grain growth inhibitors. Instead, the insoluble dispersoid particles were swept before the advancing TLP and clustered near the triple points.[3] The partially undissolved TLP contained mostly Al-Cu-Zn. Zinc in 7475 may have reacted with the Cu-rich TLP to form a low temperature ternary TLP phase, creating localized liquated region at the grain boundary. The combined effects of high volume fraction of the incoherent phases acting as the microvoid initiators [18] and TLP coating the grain boundaries have contributed to the lower strengths of the DB 7475 with the Cu interalyer.

Shear Properties of DB 8090 and 7475

The shear strength of the bond was qualitatively estimated using a theoretical relation,[19] $\tau=0.6\sigma_L$, where σ_L is longitudinal tensile strength and listed in Table 1. It is difficult to make a direct comparison between the present results and others due to the fact that many reported data are in the post-DB T6 condition. With the post-DB heat treatment, the Cu-rich phases seen in the grain boundaries of the DB joints of both 8090 and 7475 should dissolve and reprecipitate in the matrix, thereby improving the joint strength. In addition, the joints are produced in vacuum with much longer time than the ones used in this work. Despite these differences, the estimated shear strengths of 120 and 105 MPa for the Cu interlayer specimens of 8090 and 7475, respectively, are comparable to the results reported by Dunford and Partridge,[6] Pilling and Ridley, [8] and Kennedy.[9] A commercially pure Al with the Cu interlayer exhibited a maximum shear strength value of ~50 MPa after 60 min at 550°C under 3 MPa pressure in vacuum; a low strength was attributed to Al_7Cu_2Fe particles found on the bond interface.[14] The estimated shear strengths of the Zn electroplated specimens of 8090 (58 MPa) are much lower than those of the Zn physical vapor deposited specimens of 8090.[2] However, for aircraft structures, the actual shear strength requirements of the bonds are generally on the order of 10-20 MPa, and the present results satisfied the requirements. The greater importance for the bond property is its resistance to peeling, particularly during the SPF process.[8]

The diffusion bonding of 95Zn-5Al clad SPF 7475 in an argon atmosphere has not been as successful. The joints were produced using a static pressure of 1.2 MPa (175 psi) at 515°C for 60 min. The shear strength of 10 MPa was obtained. The corresponding fracture surface showed very limited shear-ductile tearing (Figure 6), while oxidation continued to be a problem. Although Zn appeared as an ideal choice for an interlayer since it forms a low melting TLP and has an excellent metallurgical compatibility with Al, however, the DB of 7475 with the Zn interlayer continues to be unsuccessful. A further study is needed to better understand the effect of Zn on diffusion bonding of Al alloys.

Conclusions

Diffusion bonding of aluminum alloys has been successfully demonstrated using the production-feasible diffusion bonding condition. The effect of surface chemistry on the DB properties of the aluminum alloys, 8090 and 7475, was evaluated for both interlayered and bare surfaces. Two interlayer elements, copper and zinc, affected each alloy differently. With the same interlayer thickness and bonding parameters, the tensile properties of the copper interlayer specimens of 8090 and 7475 were found to be substantially better than those of the bare surface and zinc interlayer specimens. For the copper interlayer specimens of 8090 and 7475, the tensile strengths of 200 and 175 MPa were obtained, respectively. For the zinc interlayer specimens, the advantages of using a low temperature transient liquid phase bonding were not observed. A further study is needed to better understand the effect of Zn on the diffusion bonding of Al alloys.

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Table 1. Tensile and estimated shear strengths of diffusion bonded 8090 and 7475 Al alloys.

	Fracture Strength MPa	Estimated Shear Strength MPa
8090 base metal	395 ± 20	223
Cu Interlayer 8090	200 ± 7	120
Zn Interlayer 8090	96.5 ± 10	58
Cu interlayer 7475	175 ± 8	105
Zn interlayer 7475	-	-

Figure Captions

Figure 1. Optical micrographys of Al alloys shown in three orientations: (a) 8090; (b) 7475.

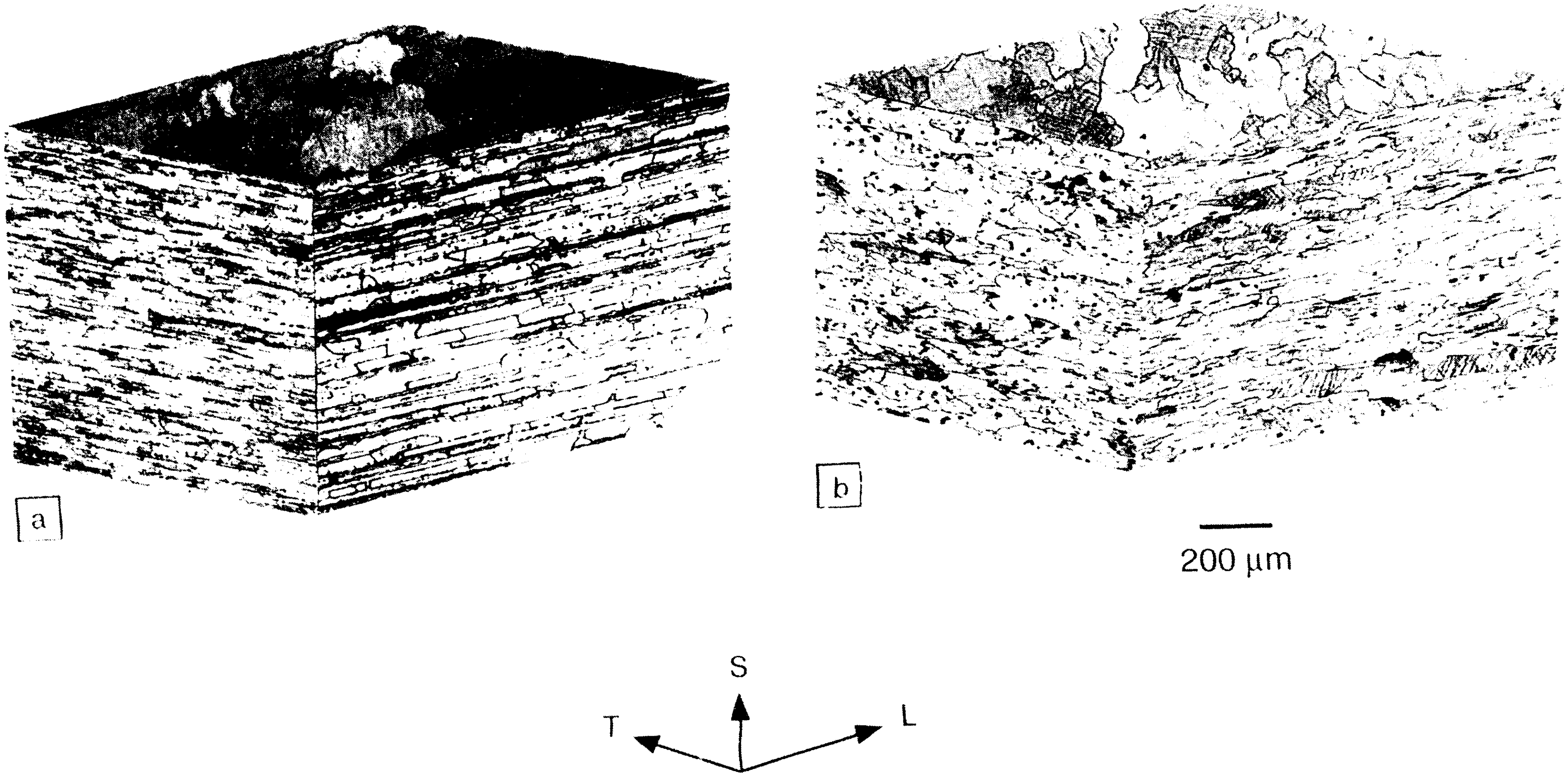
Figure 2. SEM fracture surface of diffusion bonded 8090 without an interlayer, showing the dimples formed by previously present Li-Al spinel particles on the oxided surface.

Figure 3. Cross-sctions of diffusion bonded 8090 joints showing the effect of the transient liquid phase bonding: (a) with the Cu interlayer; (b) with the Zn interlayer. The interface is indicated by the arrows.

Figure 4. SEM fracture surfaces of diffusion bonded 8090 joints: (a) with the Cu interlayer; (b) with the Zn interlayer.

Figure 5. Diffusion bonded 7475 joint with the Cu interlayer: (a) cross-section; (b) fracture surface. The interface is indicated by the arrows.

Figure 6. SEM fracture surface of diffusion bonded SPF 7475 joint with the Zn interlayer, tested in shear.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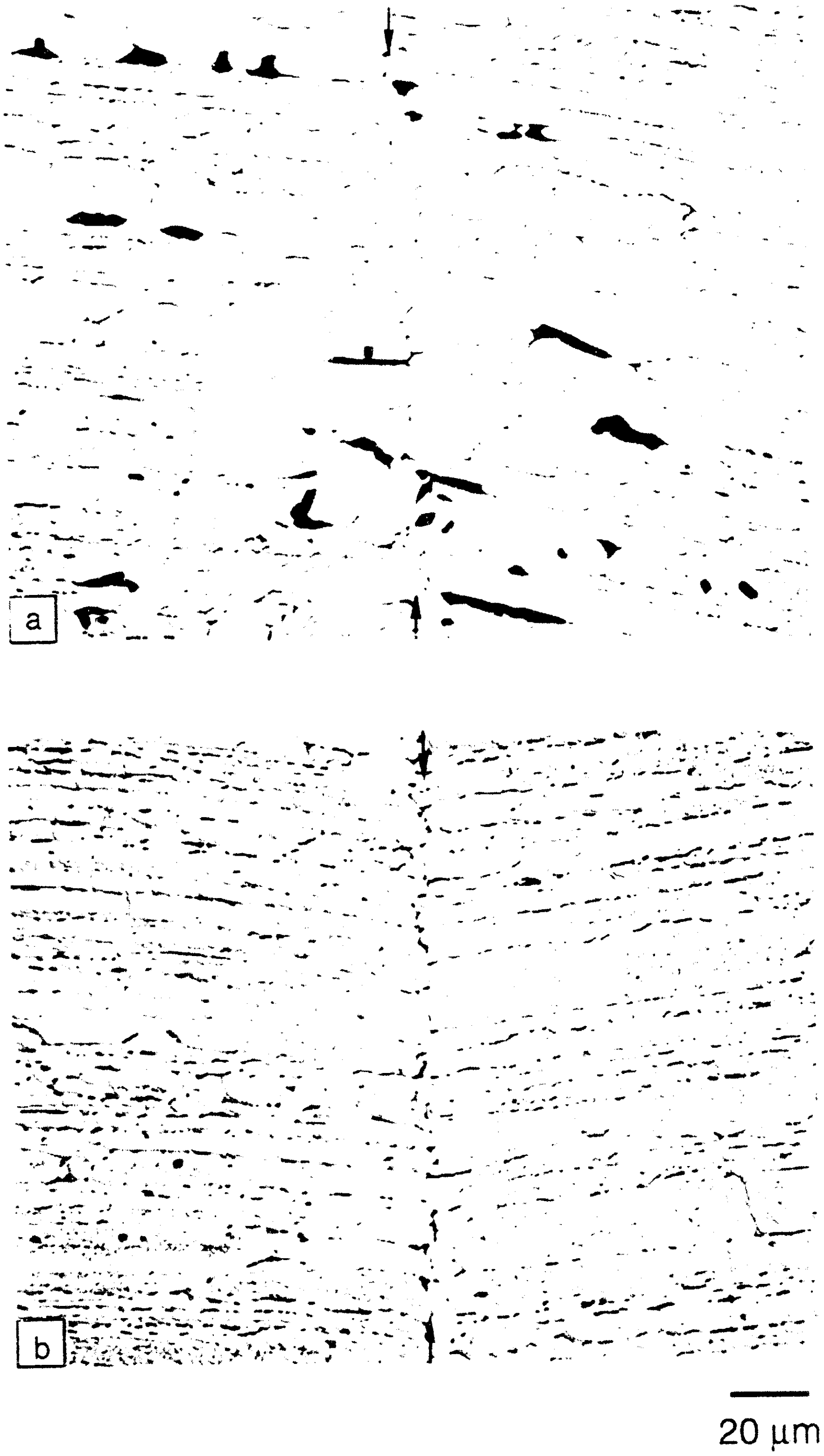
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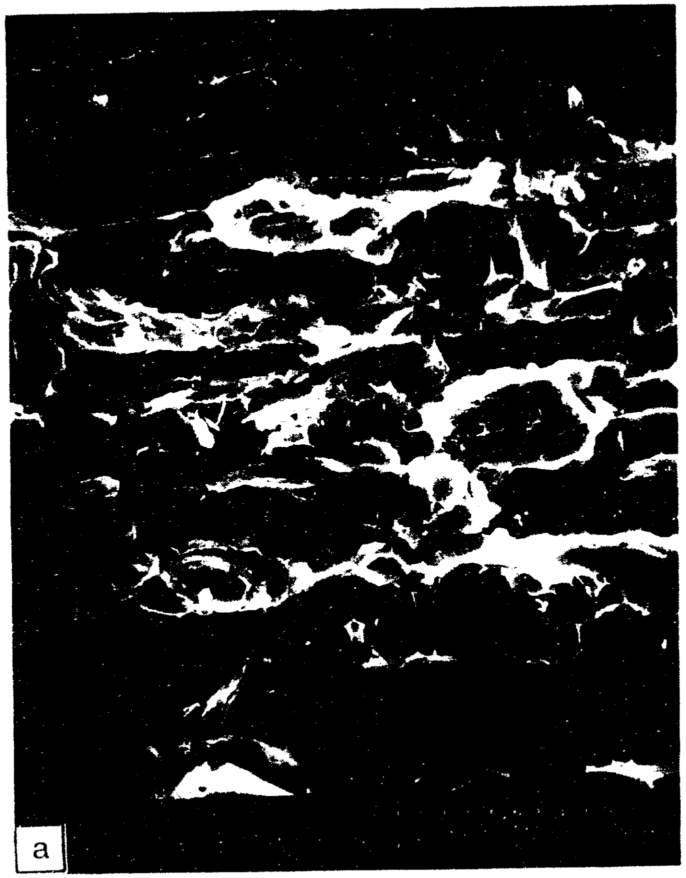
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b
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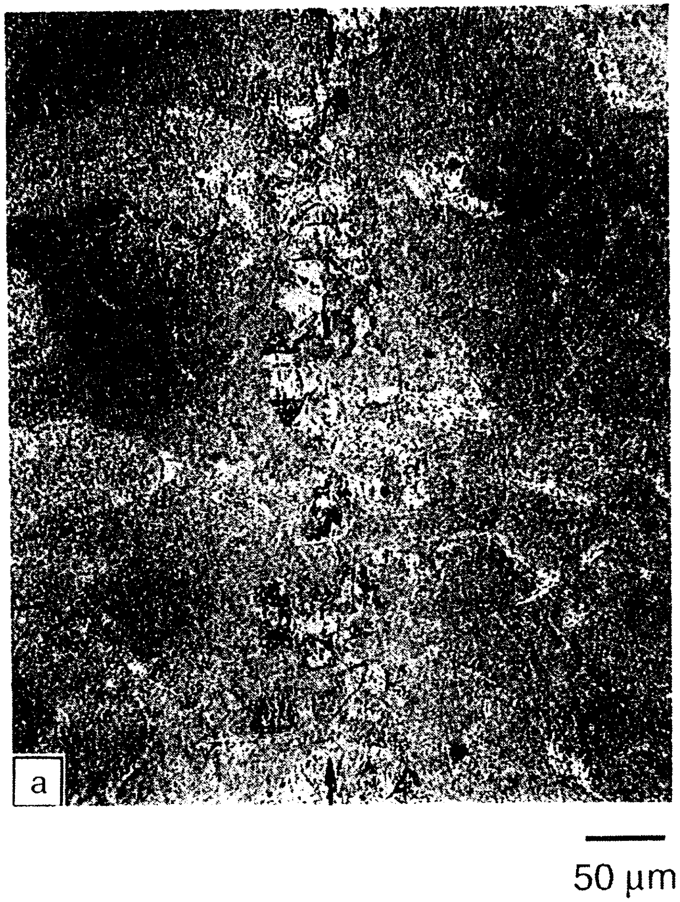
a
50 μm


7475Cu2
b
20 μm


7475 ZN-SLST #2
611054 14KV X1.50K 20.0um

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