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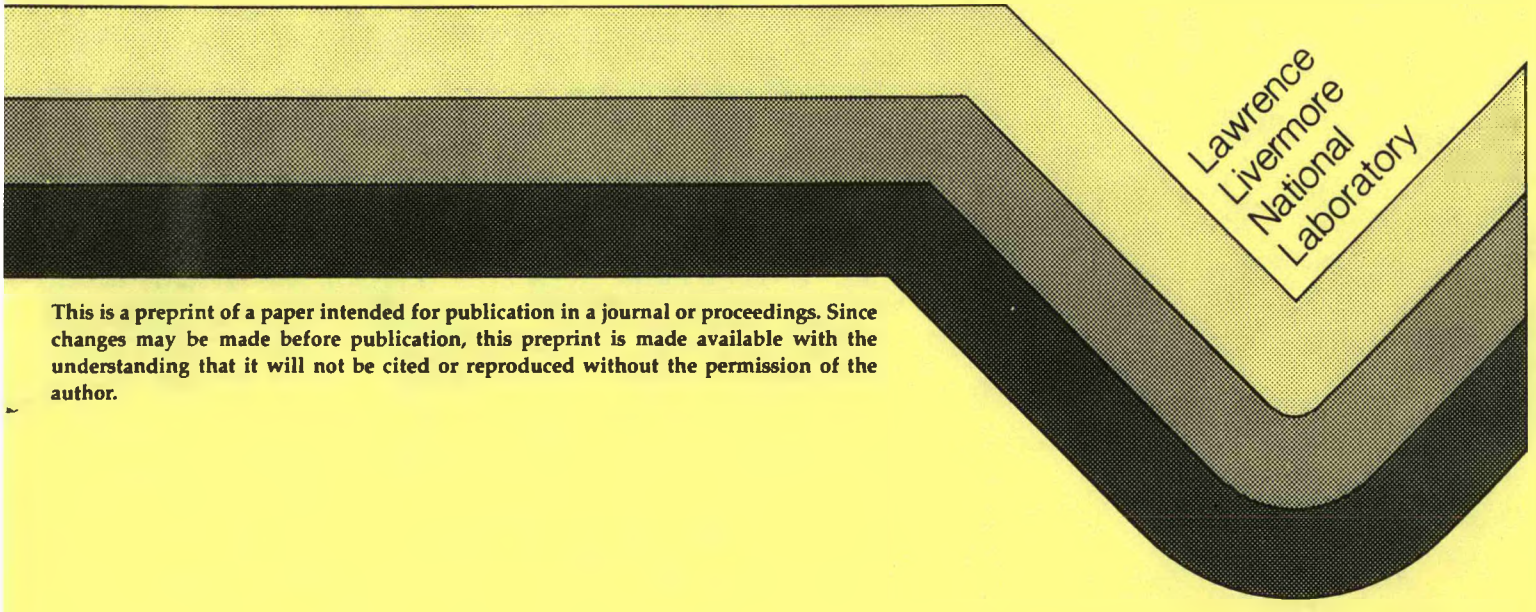

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TIME-DEPENDENT FAILURE OF SILVER-INTERLAYER DIFFUSION BONDS BETWEEN NON-DEFORMING BASE-METALS

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ABSTRACT

Silver-interlayer diffusion-bonds, fabricated at low temperatures using planar-magnetron sputtering, exhibit very high tensile strengths. Earlier work has shown that these joints undergo delayed failure at relatively low tensile stresses at ambient temperature for the case in which plasticity occurs in the base materials. Failure apparently occurs by a microvoid coalescence mechanism at the bond interfaces. Delayed tensile failures were investigated in this study for the case in which the applied stress does not produce any plastic deformation in the base metal. Failure occurs and appears to be controlled by time-dependent plasticity within the silver interlayer, which is governed by the effective stress in the interlayer. The plasticity causes cavity nucleation and, eventually, interlinkage and failure. These findings are believed to be generally applicable to any thin interlayer bond, including those prepared by processes different than physical vapor-deposition.

1. INTRODUCTION

It has long been known that thin (e.g., 1 μm - 1 mm) interlayer welds, bonds or brazes between higher strength base materials may have high ultimate tensile or rupture strengths despite the relatively low strength of the filler metal. The high strength of the joint is due to the mechanical constraint provided by the non-deforming base metals which restricts transverse contraction of the interlayer. The constraint produces a triaxial state of stress and reduces the effective stress, thus reducing the tendency for the interlayer to plastically deform [1-4]. The degree of mechanical constraint in the joint is generally known to increase with decreasing thickness-to-diameter ratio of the interlayer [5-8]. Other factors, such as plasticity of the base metal, reduce the constraint [8]. Higher joint strength is associated with higher constraint.

Recent work by the authors [9,10] has verified an earlier observation [11] of delayed or time-dependent tensile failure of solid-state interlayer bonds (of various composition) at ambient and elevated temperatures at stresses substantially less than the ultimate tensile strength. In those studies the solid-state bonds were prepared by the hot-hollow-cathode vapor-deposition process [9,10] and by interlayer foils [11]. The preliminary results [9,10] indicated that the ambient temperature time-dependent plasticity, or creep, of the base metals (e.g. to plastic strains of about 0.01 or less) relieves the constraint and the joint strength may be, correspondingly, degraded. That is, base-metal creep induces concomitant shear within the interlayer under a state of high triaxial stress which causes ductile failure within the interlayer. Failure occurs at the center-plane of the interlayer or the base-metal/interlayer interfaces after relatively small plastic strains. Therefore, the creep rate of the base-metal controls the time-to-failure.

The delayed failure phenomenon for the general case of interlayer bonds utilizing base metals that deform only elastically was not previously investigated. This question is particularly relevant since many alloys, ceramics and composites fall within this category. In the present study, ambient and near-ambient temperature creep-rupture tests were performed at a variety of stresses below the ultimate tensile strength of the bond using base metals which did not undergo plastic deformation. Interlayer bonds prepared by planar-magnetron sputtering were preferred in this study for several reasons. First, as discussed in a separate article by two of the authors [12], these interlayers have fewer impurities and voids, and have better adhesion than interlayers prepared by other methods. Also, this modern low-temperature joining process is increasingly utilized for joining ceramic and composite materials.

2. EXPERIMENTAL PROCEDURES

Silver-aided diffusion welds were fabricated using maraging steel (0.2% offset-yield stress of about 1515 MPa). The maraging steel exhibits only elastic deflections over the relevant range of applied stress (less than 760 MPa). The base-metal blanks were machined into right circular cylinders, 15.3 mm in diameter and 38.8 mm in length. The surfaces of the ends to be coated of some maraging specimens were machined flat (by single-point turning) to 2 μm with a surface roughness of 0.1 μm arithmetic average. Other specimens were lapped (ground) flat using 1-μm diamond paste to 0.15 μm and to a surface roughness of 0.03 μm.

150-μm-thick silver-interlayer bonds were fabricated by planar-magnetron sputtering and hot isostatic pressing (HIPing) at 400 °C and 207 MPa, as described elsewhere [12]. The deposited silver layer (prior to bonding) has been confirmed by TEM to consist of columnar grains with an average diameter of about 0.25 μm. The column axes are essentially perpendicular to the base-metal surface and nearly coincident with the <111> crystallographic direction. The grains contained numerous growth twins, typically 10-15 nm in thickness. Recrystallization of most (50%-75%) of this structure occurs during HIPing. A serrated, or wavy, high angle boundary generally remains, however, within the vicinity of the original silver surfaces. Typically, the recrystallized grains are greater than 1 mm long in the plane of the bond and encompass virtually the entire thickness of the original coating, i.e. 75 μm. The grains contained numerous annealing twins. Figure 1 illustrates light optical images at two magnifications of silver-aided diffusion bonds prepared using the above

process. Chemical (SIMS) analysis of the bonded interlayer did not reveal the presence of any significant quantity of impurities.

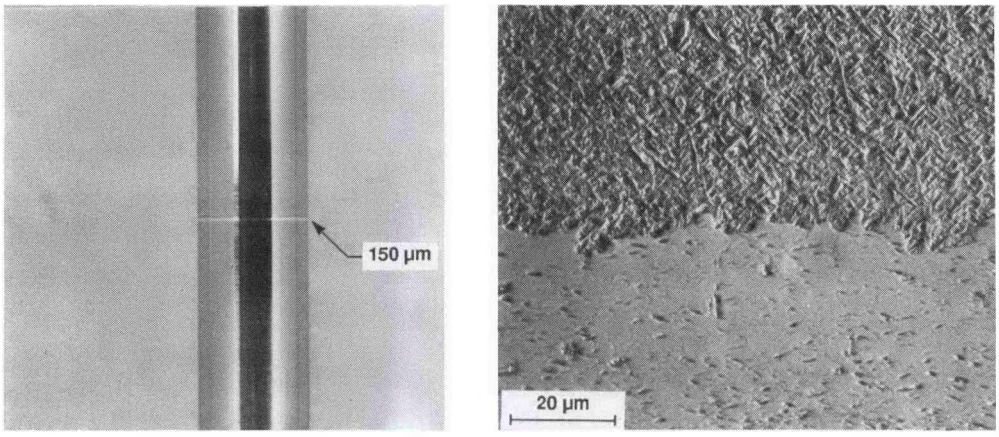

Fig. 1 (a) and (b) Optical micrographs of a silver-interlayer diffusion-weld between stainless steel. The interlayers are 150 μm in thickness.

The mechanical behavior of the bonded interlayer was determined using torsion (low mechanical constraint) tests. The 0.01% offset effective (von Mises) yield stress was measured to be about 45 MPa whereas the 0.2% offset effective yield stress was found to be about 115 MPa, indicating both a high yield strength and hardening rate compared to annealed bulk-polycrystalline-silver [13]. The higher flow stresses can be rationalized, at least partially, by incomplete recrystallization and a refined substructure in the recrystallized grains.

Tensile creep-tests were performed at ambient temperature and at 72 °C using simple lever dead-weight type creep-rupture testing machines. Heating was accomplished by resistance furnaces that controlled the temperature within ±1 °C as measured by a thermocouple spot-welded to the specimen surface. Plastic strains were measured on unloaded samples by measuring specimen diameters using an optical comparator. Tests were performed in air of 30-40% relative-humidity, and the effects of any stress-corrosion cracking were not relevant [14,15].

3. RESULTS AND DISCUSSION

3.1 Mechanical Tests

Delayed failure of the maraging-steel joints is clearly observed, as shown in Fig. 2. As the applied stress decreases, the time-to-rupture increases. Further, delayed failure is observed at stresses less than 20% of the ultimate tensile stress (approx. 700 MPa). Tests were performed at ambient temperature (circles) and 72 °C (squares). Ambient temperature tests on maraging specimens for which surfaces were lapped, rather than machined, prior to coating are indicated by triangles. At a given applied stress, lapped specimens fail at times approximately 50 times longer than those with machined base-metal surfaces. Ambient-temperature rupture-times are nearly 50 times longer than tests at 72 °C. Failure occurs at or very near one of the three principal interfaces. Fracture at the interfaces shows obvious microvoid coalescence. At

stresses between 350 and 700 MPa, the fracture surface typically consists of 85-100% silver/silver ductile separation. Below 350 MPa, the fracture surface consists typically of 55-85% silver/silver separation with the remainder of the fracture located near the silver/base-metal interface.

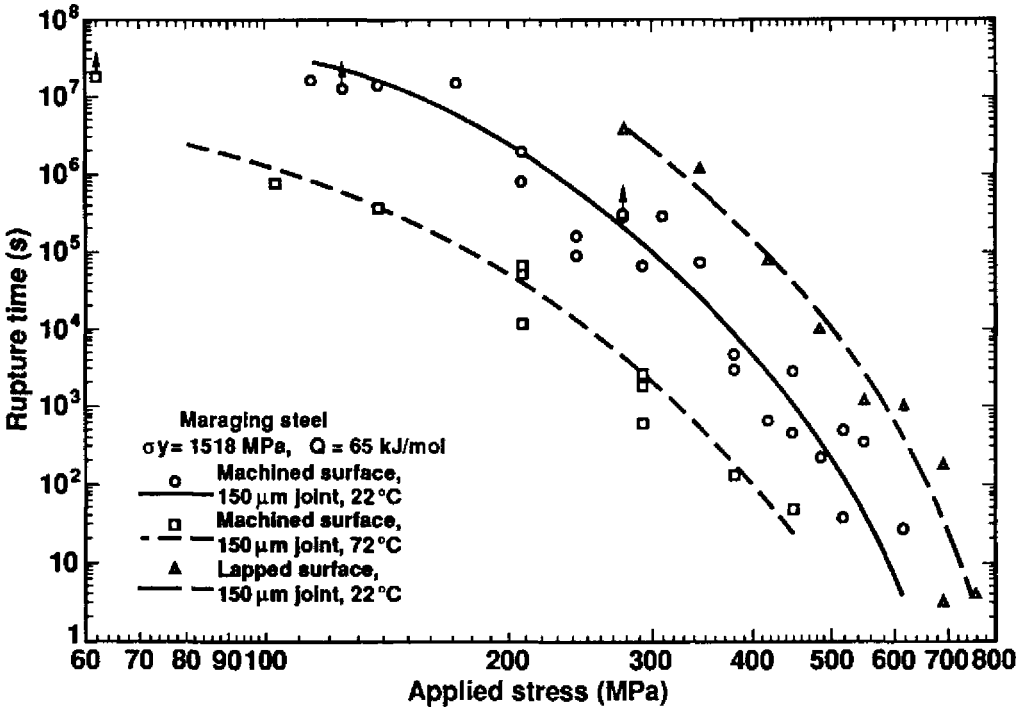

Fig. 2 The rupture-time versus applied stress for silver-aided diffusion bonds between maraging steel at ambient temperature and 72 °C. Ambient temperature tests include maraging specimens that were lapped as well as machined prior to coating. The maraging steel deforms only elastically at these stresses.

Figure 3 shows a scanning electron (SEM) micrograph of the typical silver/silver fracture surface of bonds in which the base metal surfaces were (a) machined and (b) lapped prior to coating. The silver/silver separations show classic ductile failure by microvoid coalescence. The silver/silver fracture surface of the specimen for which the base metal was machined prior to coating has a duplex fracture-morphology; relatively deep cavities are separated by regions with smaller ductile-dimples. The small dimples are roughly 1 μm in diameter, which is comparable to those of specimens in which the base metals were lapped prior to coating. This difference will be discussed more fully later in this section. The morphology of the fracture surfaces of either type of specimens does not significantly change over the very wide range of applied stress. Virtually all of the "base-metal/silver" separations are actually separations between the recrystallized interlayer silver and a thin (approx. 1 μm) unrecrystallized layer adjacent to the base metal. Thus the silver/silver fracture surfaces do not appear dramatically different from the base-metal/ silver fracture surfaces.

Delayed failure of interlayer bonds does not appear to be caused by environmentally-induced embrittlement. First, base-metal/silver delayed-failure separations appear to occur independent of base-metal and interlayer selections and the interlayer deposition processes. They have been observed at the base-metal/interlayer interfaces in austenitic and ferritic steels, U, 8091 Al alloy, Be, and Ni bonds [9,11,16,17]. Further, base-metal/interlayer separations have been observed with interlayers of varying composition and prepared using planar-magnetron sputtering, hot-hollow-cathode evaporation processes, interlayer foils, brazes and electrodeposition [9,11,16,17]. Embrittlement would not appear to be as general a phenomenon as mechanically-induced delayed-failure. Second, the temperature and environmental conditions are not expected to induce any external (e.g., environmental) cracking [14,15]. External cracks were never observed by dye-penetrant or by optical metallography prior to failure.

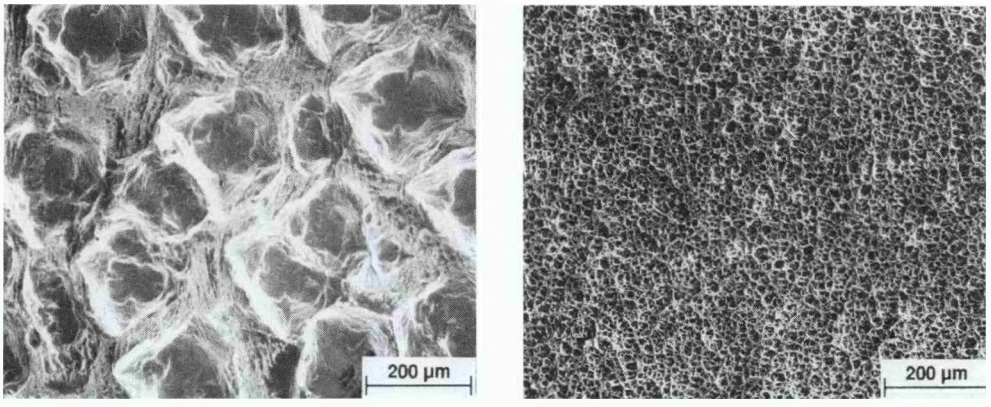

Fig. 3 The typical silver/silver SEM fracture surface for bonds in which the maraging (elastic) base metal surfaces were (a) machined (applied stress = 241) and (b) lapped prior to coating (applied stress = 758).

Instead, we believe that an attractive explanation for delayed failure is a rate-dependent mechanical process, such as creep of the silver interlayer resulting from the effective stress within the interlayer. The time-dependent plasticity can nucleate cavities that eventually lead to interlinkage and failure. Diffusive cavity-growth [18,19] does not appear tenable, since the equilibrium vacancy concentration and diffusivity at ambient temperature are too low [20] to rationalize the short failure-times at higher stresses. Since the fracture surface morphology is independent of stress (or t_r) the detailed mechanism would also appear to be independent of stress, and a diffusive (or a coupled-diffusive [18,19]) mechanism would not appear relevant at any of the stress levels that induce failure.

Careful strain measurements indicate that the "macroscopic" plastic strain-to-failure in the silver interlayer is about 0.002 at higher stresses. We postulate that the, albeit small, plastic strain on loading causes dislocation pile-ups at the three principal interfaces (as well as some other locations). A small number of

dislocations in a pile-up could be sufficient to concentrate the stress at the interfaces and nucleate a small cavity or crack, particularly in the presence of large hydrostatic tensile stresses.

The plasticity concept may also rationalize the morphology and rupture-time differences between specimens fabricated with machined and those with lapped surfaces. Silver deposition on the relatively rough machined surface results in a correspondingly rough silver surface, even after 75 μm of silver deposition. During the bonding cycle, the joining of the two rough surfaces may result in some substructural heterogeneities (but not voids) near the bondline that are less common in bonds using the smoother lapped surfaces. The heterogeneities in the machined specimens may provide for higher cavity nucleation-rates and the formation of large cavities. Fracture may be accelerated in these regions, resulting in shorter rupture times, and may occur more heterogeneously, as evidenced by the relatively large, deep, cavities.

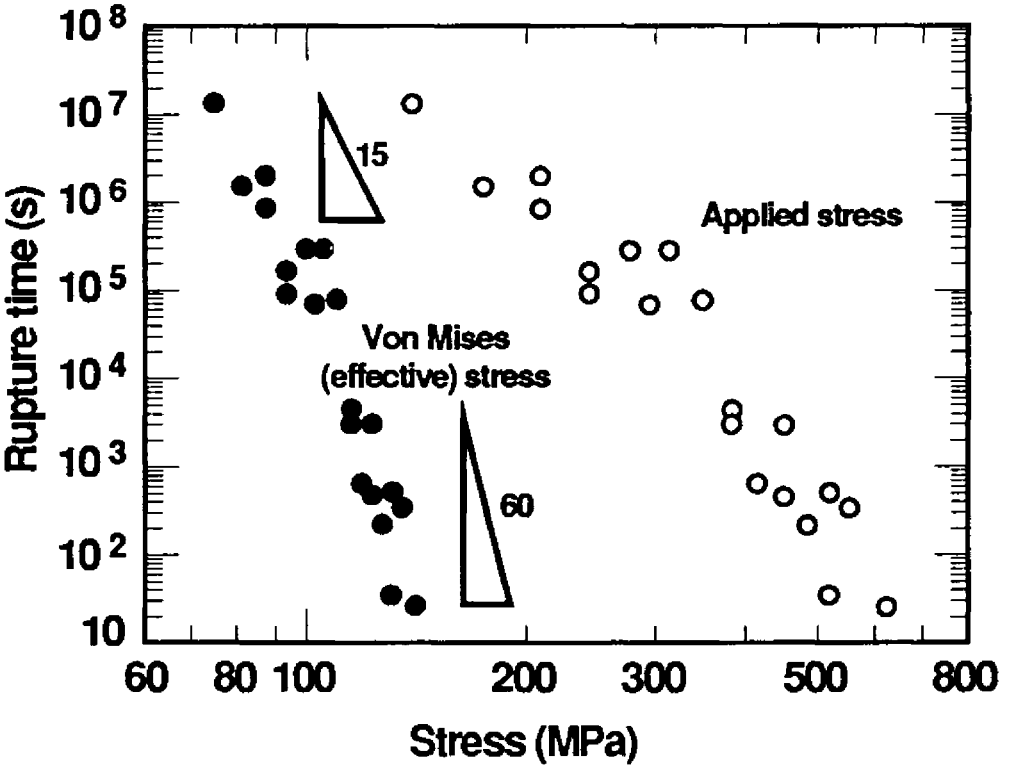

Fig. 4 The rupture-time versus applied and effective stress within the interlayer for diffusion bonds between maraging steel at ambient temperature.

Cavity interlinkage, which probably leads to the catastrophic ductile delayed failure, may occur once a critical concentration (which would be a function of the applied load) of nuclei or cavities is attained at the interface(s). The concentration of interfacial cavities is apparently time-dependent. The nucleation, growth and/or interlinkage rate is expected to be a function of the

creep rate of the silver at locations in the vicinity of the interface. Finite-element-method (FEM) stress analysis was performed on the 150-μm interlayer bonds {using the NIKE 2D code [21,22] with 2256 elements (480 in the interlayer)}. These results are shown in Fig. 4, where the data of Fig. 2 (specimens with machined base metal surfaces) are plotted as a function of the effective (von Mises) stress and the applied stress. The analysis revealed that the effective (von Mises) stress within the vicinity of any of the interfaces is above the interlayer yield-stress at all the applied loads at which delayed failure is observed. Therefore, nucleation of cavities associated with creep could occur at any of the applied stresses at which delayed failure is observed. The significance of the slopes will be discussed later.

The silver-plasticity concept for rationalizing delayed failure is consistent with the temperature dependence of the time-to-rupture. The activation energy for plastic flow, Q, is usually defined by:

$$Q = -R\left[\frac{\partial \ln \dot{\varepsilon}}{\partial \ln (1/T)}\right]_s \qquad (1)$$

where R is the gas constant, $\dot{\varepsilon}$ is the strain-rate, T is absolute temperature and s is the substructure. For creep of silver at ambient and near-ambient temperatures for various substructures, Q falls within the range of 50-71 kJ/mole [13,23]. For the delayed failure of silver-interlayer diffusion bonds using maraging steel Q is estimated from Eq. (1) by substituting $\dot{\varepsilon}$ by $1/t_r$. This estimation is valid if for a given applied stress the plastic strain-to-failure within the interlayer is approximately fixed. The resulting activation energy of 65 kJ/mole falls within the range for silver plasticity.

3.2 Microscopy

An important test for the verification of the proposed theory to explain delayed-failure is examination of the silver/silver and base-metal/silver interfaces at various fractions of the creep-life. If delayed failure is the result of silver creep under high triaxial stresses that leads to void nucleation, growth and interlinkage, then cavities should be increasingly evident at or very near the interfaces with an increasing fraction of the expected rupture-time, $t_{r,ex}$. Therefore, specimens were loaded at different stresses for various fractions of $t_{r,ex}$, unloaded and examined for cavities. Using light-optical techniques, small cavities at the three interfaces were frequently observed in specimens loaded to 10% or more of $t_{r,ex}$. Figure 5 presents optical micrographs of bonded specimens utilizing machined maraging steel that were (a) as-bonded (0%) and (b) loaded at 124 MPa for 50% of the expected rupture time. Consistent with the proposed explanation for delayed failure, numerous cavities are evident at the silver/silver interface, as well as at grain boundaries in non-recrystallized regions.

3.3 Failure Theory

The previous experimental results are substantially supportive of the hypothesis that the delayed tensile failure of thin interlayer bonds is a result of plasticity occurring in the soft interlayer under high triaxial stresses, leading to

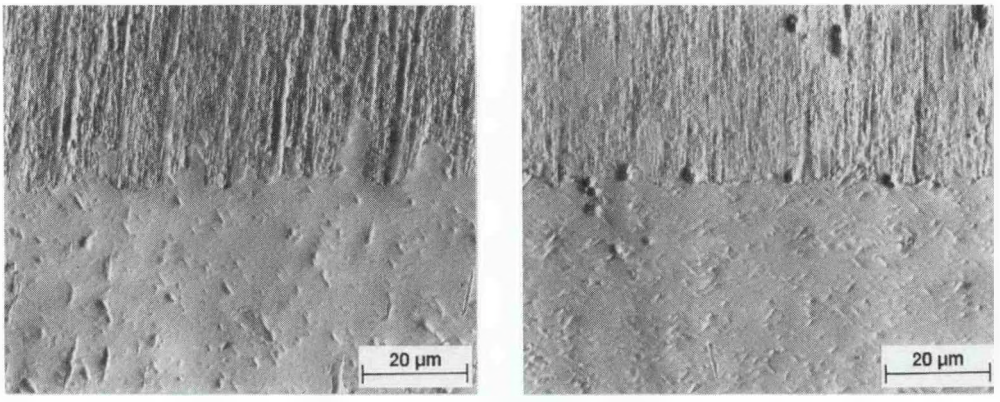

Fig. 5 Optical micrographs of the silver/silver interface in lapped specimens loaded to (a) 0%, and (b) 50% of the expected rupture time at a stress of 124 MPa .

the nucleation, growth and interlinkage of cavities. The temperature-dependence of the rupture times, FEM analysis, and optical microscopy, are all consistent with a creep-plasticity explanation for failure.

However, some questions remain. For example, if creep is associated with the rate-controlling process for failure, then the stress sensitivity of t_r should be related to the stress exponent for plasticity of silver, just as the temperature dependence was found to be related to the activation energy for silver plasticity. Two common definitions of the stress exponent are the "steady state" stress exponent, n

$$n = \left[\frac{\partial \ln \dot{\varepsilon}_{ss}}{\partial \ln \sigma_{ss}}\right]_T \qquad (2)$$

and the "constant-structure" stress exponent, N

$$N = \left[\frac{\partial \ln \dot{\varepsilon}}{\partial \ln \sigma}\right]_{T,s} \qquad (3)$$

where σ is the stress and the subscript "$_{ss}$" refers to steady state. In general, the stress exponent will be between these two limiting values. For silver deformed at ambient temperature N is about 200, whereas n is between 10 and 40 [13,23]. Torsion tests of the interlayer silver of this study showed that saturation or steady state is achieved by strains between 0.05 and 0.10. Therefore, for the silver relevant to this study, as the plastic strain approaches zero, the apparent stress exponent is about 200 and for strains near 0.05 or 0.10 [23], the apparent stress exponent is between 10 and 40.

The stress sensitivity for failure of the silver-interlayer bonds between maraging steel is given by the slope of the t_r *vs* effective stress curve shown in Fig. 4. The slope varies from about 60 at high stresses to between 10 and 15 at low stresses. These values appear roughly consistent with the established steady-state stress exponent of silver but not with the constant-structure exponent. Therefore, consistency with the silver-plasticity theory is realized provided that the plastic strain accumulated to the onset of catastrophic ductile failure is near 0.05. As mentioned earlier, the measured "macroscopic" plastic strains at the higher applied stress were only about 0.002, (perhaps even smaller at lower stresses) which is less than that which would appear to bring consistency. However, if the plastic deformation in the vicinity of the interfaces is higher than the macroscopic strain (e.g., a few percent, which we believe is possible), consistency is realized.

It is also difficult to explain the apparent cavity growth. As illustrated in the previous figures, cavities with diameters between 0.2 and 1 μm are frequently observed (it is assumed that this estimate is not dramatically affected by an apparent increase in cavity size by chemical etching). If dislocation pile-ups are responsible for cavity nucleation, then the nuclei size would be on the order of a nm or so. An increase of 2 or 3 orders of magnitude in size cannot be readily rationalized by vacancy accumulation. Furthermore, this increase is not easily explained by "uniform" plastic deformation of the interlayer matrix. There is not obvious evidence that a sufficient strain level exists in the vicinity of the interfaces.

One possible explanation for the "expansion" of cavities is the interlinkage of small nuclei or cavities. On loading of an interface, dislocation activity may be most pronounced in certain regions. Small cavities may nucleate in groups in which the spacing may be small and interlinkage is possible by plasticity in the region between the small nuclei. Thus "growth" or cavity expansion may occur by interlinkage (aided by the increased effective stress between the voids) rather than the traditional concept of a uniformly expanding cavity surface. Therefore, the failure steps may be reasonably described by cavity nucleation and interlinkage rather than nucleation, growth and interlinkage. This suggestion of a non-growth control for failure has been, of course, suggested in other, simpler, systems such as uniaxial-creep [25]. Our suggestion is speculative and is an important new area for further investigation.

4. CONCLUSIONS

Thin silver-interlayer diffusion bonds between maraging steel base metals were prepared using planar-magnetron sputtering. The maraging steel exhibits only elastic strain over the range of applied tensile stress. The following conclusions are based on experiments in which the bonded specimens were loaded to various stresses below the ultimate tensile strength.

1. Delayed tensile-failure was observed at stresses as low as 17% of the ultimate tensile strength of the bonds.
2. The time-to-failure is controlled by the creep rate of silver near the interfaces in the interlayer, which is determined by the effective stress within the interlayer.
3. This model was substantiated by careful analyses of the stress and temperature dependence of the rupture-times, finite-element-analysis of the

stress state within the interlayer, as well as microscopy of the interfaces loaded to a fraction of the expected rupture-time.

4. These findings are believed to be applicable to bonds in which the interlayers are prepared by physical vapor deposition and other processes, such as brazing, electroplating, solid foils etc.

Acknowledgment

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