Ductility of a dental Ag-Pd-Cu-Au alloy

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A silver-palladium type of dental alloy for fixed restorations has been investigated with regard to the ability of the material to be plastically deformed in uniaxial tensile testing after two different age-hardening treatments. A certain degree of ductility is required for the burnishing of the margins. Aged to peak hardness at 350°C after solid-solution annealing at 900°C, the material was found to be brittle owing to a reaction zone along the grain boundaries, promoting an intergranular fracture. When the age-hardening temperature was lowered to 275°C, a less pronounced reaction zone along the grain boundaries could be observed. The accompanying mechanical properties after precipitation hardening at 275°C are probably an acceptable compromise between mechanical strength and ductility. Small particles along grain boundaries and brittleness were also found after solid-solution annealing at 900°C and quenching. It is suggested that the improved ductility after subsequent aging at 275°C is due to a coarsening of these small particles. In the as-cast condition the alloy was softer and more ductile than in the age-hardened state. □ *Age-hardening; elongation; grain boundary failures; tensile testing*

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The Ag-Pd-Cu-Au type of dental alloys has been in use for fixed restorations for several years, because of the low price as compared with gold-based alloys, the reasonably good castability, and corrosion properties that in many cases can be accepted. Another favorable feature of this type of alloy is its ability to be age-hardened in the temperature range 250–400°C owing to precipitation of ordered PdCu particles (1, 2). A further requirement that so far has received little attention in the literature is the ability of a casting of this type of alloy to be cold-formed so that its margins closely adapt to the finish line of the tooth.

The aim of the present work was to study the ductility of a commercial Ag-Pd-Cu-Au alloy annealed to two yield stress levels in addition to the as-cast state by measuring the strain to maximum load or fracture in uniaxial tensile tests. The purpose has further been to delineate the structural reasons for different ductilities.

Materials and methods

The composition of the alloy (Hvitstöp, K.A. Rasmussen A.S., Norway) was determined by X-ray fluorescence and found to be as follows in wt %: Ag, 55.4; Pd, 24.5; Cu, 13.5; Au, 5.3; and Si, 0.5.

The procedure for melting and casting of tensile samples has been described previously (3). Argon gas was used to protect the melt against oxidation.

The tensile samples had a diameter of 2.9 mm, and the initial length of the strain gauge was 11 mm. The ram speed during tensile testing (Wolpert Testatron) was 0.5 mm/min.

Heat treatments of the specimens in evacuated quartz ampullae and the results of the tensile tests are summarized in Table 1. The alloy melts at 970°C according to information from the manufacturer, and a solid-solution annealing temperature of 900°C (7) should therefore be sufficiently

Table 1. Mechanical properties

Conditions	Parallel no.	$\sigma_{0.05}$, MN/m^2	$\sigma_{0.05}$, MN/m^2	$\sigma_{0.2}$, MN/m^2	$\sigma_{0.2}$, MN/m^2	UTS, MN/m^2	UTS, MN/m^2	e_{tot}*, %	e_{tot}*, %	e_{unif}, %	e_{unif}, %	HV5
As-cast, aircooled in the investment	1	361		401		502		18.6		13		
	2	362	350 s.d. 20.5	387	403 s.d. 17.1	483	497 s.d. 12.7	18.8	20.1 s.d. 2.4	13	13.3 s.d. 0.6	169
	3	326		421		507		22.8		14		
900°C 15 min, quenching, 275°C 8 h	1	489		538		609		11.1				
	2	576	537 s.d. 44.2	635	588 s.d. 48.5	701	656 s.d. 46.0	4.9	7.6 s.d. 3.2			238
	3	546		590		657		6.7				
900°C 10 min, quenching, 350° 30 min	1	580		667		799		4.3				
	2	533	585 s.d. 54.2	656	669 s.d. 14.6	683	727 s.d. 63.1	1.0	2.1 s.d. 1.9			257
	3	641		685		698		1.1				

* Measured from the load-elongation curves. The elastic strains are not subtracted.

below the melting point. Three parallels were tested for each specimen condition. The average Vickers hardness number from five indentations on each sample with a 5-kg load (HV5) are included in Table 1.

The annealed structures were studied metallographically by sectioning the fractured tensile samples longitudinally. Polishing was carried out by using diamond paste down to 1 μm and etching by applying 2.5% KCN and 2.5% $(NH_4)_2S_2O_8$ in water for 10 sec.

The sectioned surfaces were studied by means of standard light optical microscopy and scanning electron microscopy (SEM) (Jeol). The grain size was measured by the linear intercept method. The fracture surface of tensile specimens was examined by SEM.

Results

The mechanical properties of the tensile tests are given in Table 1. It can be seen that age-hardening causes a significant increase in yield strength, ultimate tensile strength, and hardness but at the same time a lowering of the total elongation. These differences in properties are illustrated in Fig. 1 by a representative load-elongation curve from each group of materials Particularly, the highest

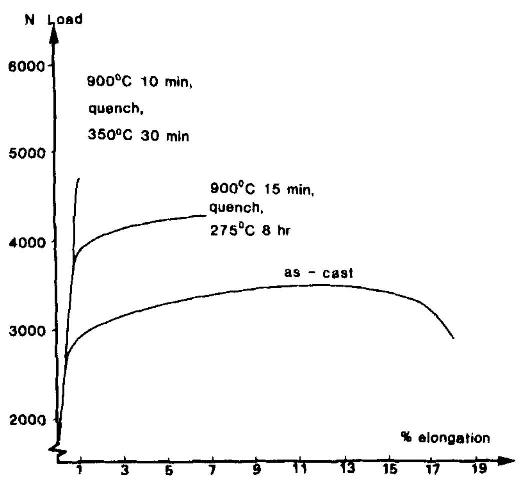


Fig. 1. Tensile curves for the three investigated conditions of the alloy.

Fig. 2. Micrograph of the as-cast structure. Etched. (×130.)

age-hardening temperature (350°C) caused a low ductility. In addition, it was observed that specimens quenched from 900°C without any further heat treatment were so brittle that they fractured merely by being dropped on the floor.

In the as-cast condition the microstructure is dendritic, as shown in Fig. 2. An almost homogeneous solid solution is obtained by annealing at 900°C, but in spite of quenching

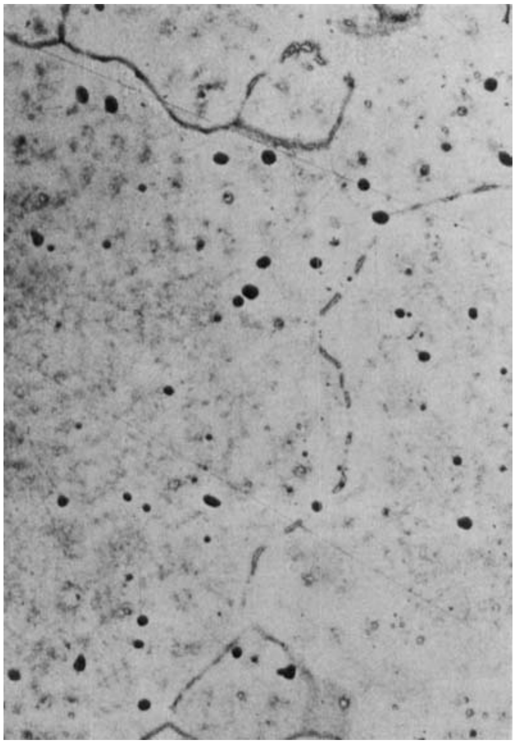
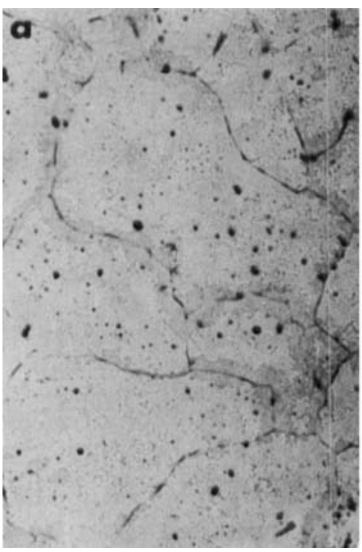


Fig. 3. Micrograph of the structure after solid-solution annealing at 900°C for 15 min, quenching, and aging at 275°C 8 h. Etched. (×130.)



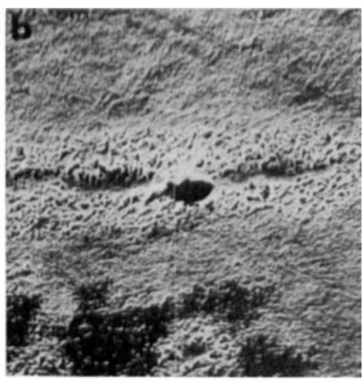
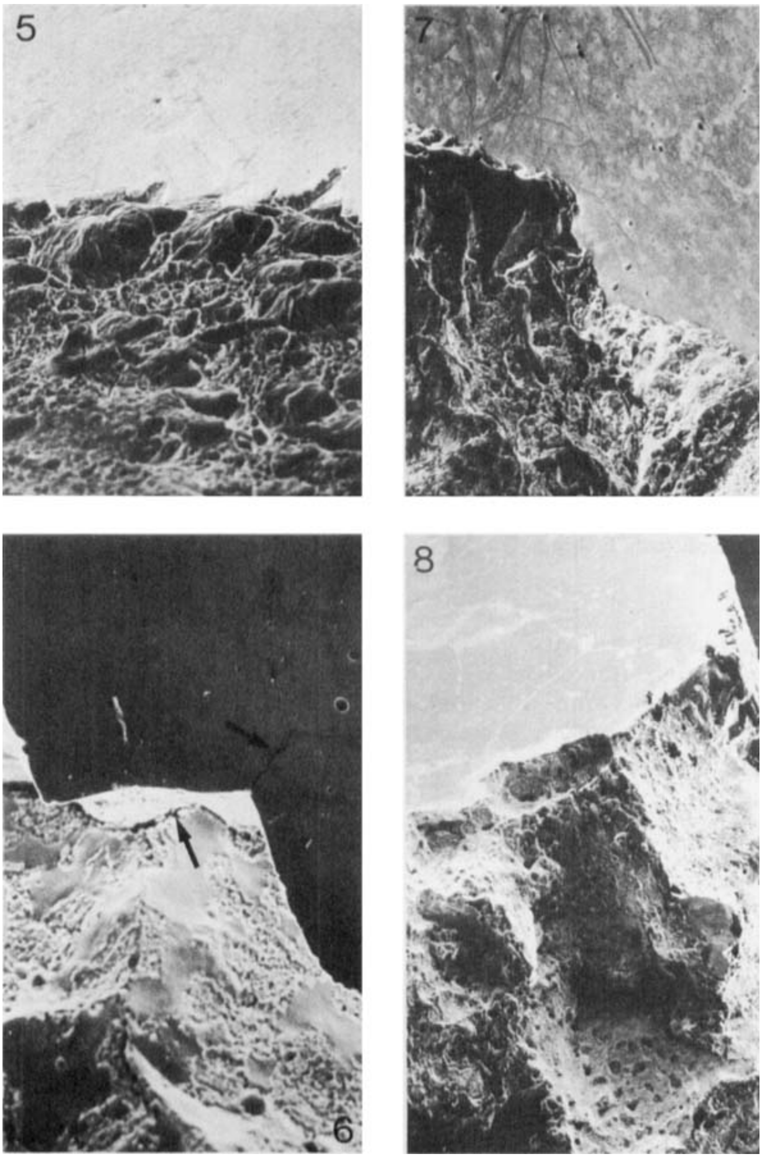


Fig. 4. Micrographs of the structure after solid-solution annealing at 900°C for 10 min, quenching, and aging at 350°C 30 min. Etched. 4a. Light optical microscopy (×130). 4b. SEM (×3000).



Figs. 5, 6, 7, and 8. SEM two-face views of fractured tensile samples, showing both the polished/etched longitudinal cross-section and the fracture surface.

Fig. 5. As-cast. (×500.)

Fig. 6. Annealed at 900°C for 15 min in a quartz ampulla and quenched. Arrows indicate small particles along grain boundaries in the polished section and intergranular cracks in the fracture surface. (×300.)

Fig. 7. Aged at 275°C for 8 h. (×100.)

Fig. 8. Aged at 350°C for 30 min. (×100.)

by fracturing the ampullae in water, some small particles were precipitated along grain boundaries. Subsequent age-hardening at 275°C and 350°C produced more extensive reaction zones along grain boundaries, as can be seen from the micrographs in Figs. 3 and 4. These structural features can also be recognized in the SEM micrographs of the fractured tensile samples in Figs. 5, 6, 7, and 8.

Discussion

It has previously been found that peak hardness in an alloy with a composition similar to the present one is achieved after solid-solution annealing at 900°C, quenching, and subsequent age-hardening at 350°C to 400°C (2, 7). The results of the present tensile tests on samples heat-treated in this manner, however, show that the material is brittle. In two of the three tensile specimens there was hardly any plastic deformation at all before fracture (Table 1, Fig. 1). The SEM micrograph with a two-face view of the fractured specimen (Fig. 8) clearly demonstrates that the failure occurs along the grain boundaries. The specimen was quite course-grained (240 μm), as was all the examined material after annealing at 900°C. The polished and etched section in Fig. 8 indicates a reaction zone along and adjacent to the grain boundaries, which is even more clearly illustrated in Fig. 4a. At a higher magnification precipitated particles can be observed (Fig. 4b). It is well known that such particles can make the material prone to intergranular fracture. The structure of the discontinuous particles at grain boundaries after annealing at 350°C in an alloy similar to the present one has previously been found by transmission electron microscopy studies to be complex (1). These investigations indicated that the particles consisted of alternating lamellae of a CsCl type of structure and Ag-rich f.c.c. material. Zones with particles of this kind have previously been reported by Ohta et al. (2) at temperatures above 275°C and up to 375°C. The lowest temperature for their occurrence depended on alloy composition and grain size. Precipitation of hardening particles (PdCu-ordered f.c.t.) took place at a wider temperature range in the interior of the grains. Their findings are in accordance with the present observations, as age-hardening at 275°C was found to cause a much narrower reaction zone along the grain boundaries (Fig. 3) and a more ductile-like appearance of the fracture surface with tendencies to dimples (Fig. 7).

The smaller reaction zone along the grain boundaries is thus a probable reason for the substantially improved ductility with an average elongation of 7.6%, compared with 2.1 for the specimens aged at 350°C (Table 1). However, it should be noted that the load-elongation curves for the material age-hardened at 275°C did not pass through a maximum (Fig. 1), so that the total elongation equals the uniform elongation for this material. This indicates a tendency to brittleness even for this age-hardening condition.

Furthermore, an extreme brittleness and intergranular fracture (Fig. 6) were observed even in the quenched specimens after annealing at 900°C in quartz ampullae and before aging. Small particles have been precipitated along grain boundaries either at 900°C or during quenching, as can be seen on the polished section in Fig. 6. It is well known that even thin films may cause intergranular fracture and brittleness. It can be suggested that upon subsequent aging at 275°C the improved ductility is due to an agglomeration of the initial film or particles, in addition to further precipitation and growth of particles. At 350°C the amount of grain boundary particles becomes so large that the ductility decreases again. Both the polished sections and the fracture surfaces in Figs. 6, 7, and 8 support this suggestion.

The material aged at 270°C also had a larger grain size (560 μm) than the samples aged at 350°C (240 μm) (Figs. 3 and 4), probably due to a slightly extended holding time at 900°C (Table 1). No grain growth due to lowering of the grain boundary energy can be expected at temperatures up to 350°C in this alloy. The movement of grain boundaries requires diffusion, which increases significantly at temperatures above approximately half of the homologous melting point. The increased grain size, however, probably has an adverse effect on ductility. The reason for this assumption is that a crack can be nucleated from stress concentrations at the head of a pile-up of dislocations against a grain boundary. This stress concentration has been calculated to increase with the square root of the grain size (5). Furthermore, such a relationship has been observed for cleavage fracture in steels. The increased grain size in the material aged at 275°C is therefore an unlikely explanation for the increased ductility. This improved property, however, was achieved at the expense of a lowered mechanical strength. The yield stress $\sigma_{0.2}$ dropped from 669 MN/m^2 (350°C) to 588 MN/m^2 (Table 1).

The as-cast material had an even weaker mechanical strength with $\sigma_{0.2} = 403$ MN/m^2 and at the same time a high total elongation of 20% (Table 1) and a fracture surface with many dimples (Fig. 5), which is normally associated with a ductile failure. The uniform elongation, which equals the strain to maximum load, was 13% and thus shorter than the total elongation. It is unknown whether the post-uniform elongation reflects significant ductility in connection with burnishing. Localized necking with high hydrostatic tensile stresses occurs after maximum load has been achieved (6) while burnishing is carried out in a manner that must be expected to create high shear stresses and weaker tensile stresses. In future work it could therefore be worthwhile trying a torsion test in which such a stress system prevails as a mean of measuring the ability of a material to be burnished.

The stress systems during uniform elongation are also different from those that are present in burnishing. Baran & Woodland (7) have loosely estimated a minimum of about 5% elongation in uniaxial tension to be a requirement for burnishing to be successful. If this assumption is correct, solid-solution annealing at 900 °C and subsequent age-hardening at 275 °C therefore seem to create mechanical properties with an acceptable compromise between strength and ductility for the present alloy.

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