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ORIGIN OF THE WIDMANSTATTEN MICROSTRUCTURE IN HOMOGENIZED Ag-Sn DENTAL ALLOYS+)

Harsono Wirjosumarto*)

Richard S. Mateer**)

R I N G K A S A N

Struktur Widmanstatten yang timbul pada paduan logam dental Ag-Sn disebabkan oleh proses pendinginan lambat paduan tersebut dari suhu homogenisasi. Hal ini terjadi bila paduan tersebut mengandung perak atau ekivalen perak antara 75 - 80 % berat.

Dalam proses preparasi spesimen untuk penelitian metalografi, garis-garis serupa dengan struktur Widmanstatten dapat timbul. Garis-garis ini sebenarnya adalah struktur kembar mekanis (mechanical twins) yang terjadi karena deformasi plastik pada proses penggerindaan dan pemolesan.

A B S T R A C T

Widmanstatten microstructures are often observed in Ag-Sn dental alloys, after they underwent slow cooling from homogenization

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*) Department of Mechanical Engineering, Institute of Technology, Bandung, Indonesia.

**) College of Engineering, University of Kentucky, U.S.A.

temperature. This structure appears when the alloy contains silver or silver equivalent between 75 and 80 wt %.

Hatchet mark structure somewhat similar to Widmanstatten structure may be generated during the metallographic preparation of the specimens. These marks are actually mechanical twins resulted from the plastic deformation during the process of grinding and polising.

INTRODUCTION

Properties of dental amalgam alloys as any other materials, depend on their compositions and the microstructure into which they form.

Dental amalgam alloys are generally considered to consist of Ag_3Sn or γ phase, although regions of "basket-weave pattern" or Widmanstatten microstructure are often observed in commercial chip alloy[1]. A search of the literature revealed that Petrenko[2] and Murphy[3], on their work in determining the phase diagram of Ag-Sn binary system, had observed such structure near the Ag_3Sn or β phase and no explanation for the structure was offered. Murphy observed the structure and suggested that it resulted from rejection of γ on clevage planes of β during slow cooling. This logical interpretation was, however, confused by occurrence of somewhat similar hatchet markings for compositions both higher and lower in silver. Murphy believed these to be homogeneous and could not explain the markings.

The present metallographic study was undertaken to test Murphy's conclusions regarding γ rejection on cooling of β to form Widmanstatten, and to explain the origin and nature of the hatchet markings. It is expected that the results of this study could lead to systematic development of improved silver-tin dental amalgam alloys.

MATERIALS AND METHODS

1. *Materials and Material preparations*

Three binary alloys of Ag-Sn system of 26.5 wt % Sn, 23.0 wt % Sn and 20.0 wt % Sn were prepared by resistance melting of 99.9 % purity Ag with 99.993 % purity Sn in graphite crucible and centrifugal casting into Vycor tubing to yield ingot with 5 mm in diameter and 50 mm in length. Tops and bottoms of cast and of homogenized ingots were examined in longitudinal and transverse metallographic section for structural uniformity. The silver and tin contents were determined by wet chemical analyses[(4)].

Commercial dental amalgam chip alloys were also examined in as received condition and after homogenization treatment.

2. *Heat Treatments*

Both the laboratory made and the commercial alloy specimens were sealed, under vacuum, in pyrex tubing and held for 48 hours at 400°C to homogenize their structure. One series of capsules were quenched into water to secure high cooling rate and a duplicate series was furnace cooled.

3. *Metallographic preparation*

The preparation of specimens for metallographic study was done according to the conventional methods, e.g.: setting each sample in cold moulded plastic followed by grinding, mechanical polishing and manual etching operations. The etching reagent used in this work was the modified Crowell's etchant*).

RESULTS AND DISCUSSIONS

1. *Ag-Sn Phase Diagram*

The phase diagram for Ag-Sn system, which is currently accepted, is the one proposed by Murphy[(3)] in 1926, which is shown in Figure 1. This diagram is used for explaining the microstructures ovserved in this work. The

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*) Modified Crowell's etchant:

$K_2Cr_2O_7$	9 grams
H_2SO_4	36 cc
NaF (sat. solution)	18 cc
H_2O	450 cc

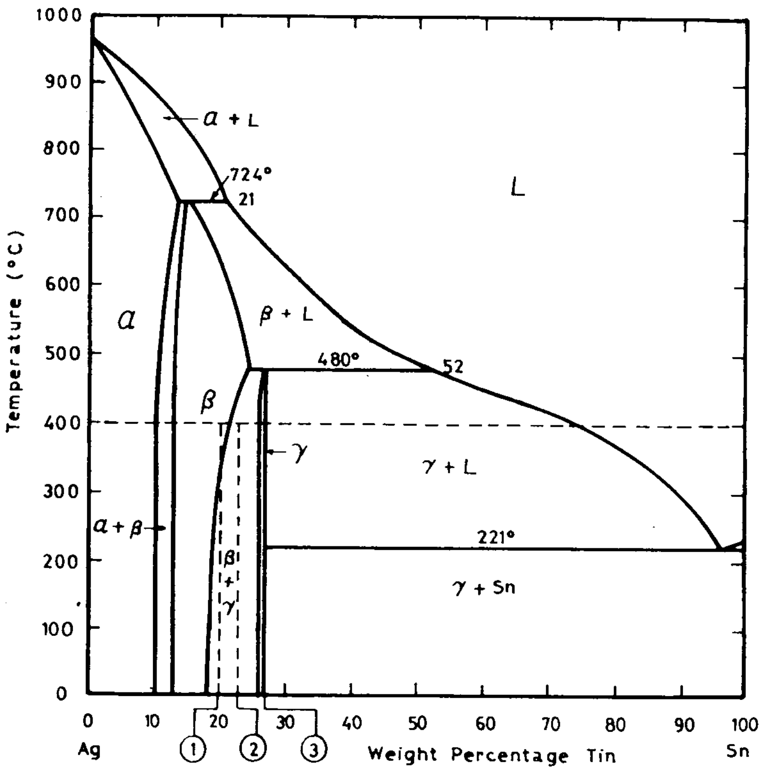

Figure 1. Phase diagram of Ag-Sn system

Composition 1 is 20.0 wt % Sn alloy
Composition 2 is 23.0 wt % Sn alloy
Composition 3 is 26.5 wt % Sn alloy

composition of the laboratory cast ingots are indicated in this diagram.

In as cast condition, all specimens showed complex microstructures and shringkage porosity associated with the peritectic solidification. By homogenization treatment, simpler structure was obtained.

2. *γ Phase or Ag_3Sn and the Hatchet Marks*

The 26.5 wt % Sn alloy, intended as stochiometric Ag_3Sn or γ phase, was uniform in structural appearance, regardless of cooling rate from the homogenizing temperature. The microstructures of the slow cooled and fast cooled sample are shown in Figure 2 and 3 respectively. Structure in Figure 2, reveals an array of "tire tread" or "hatchet mark" patterns. These marks, however, can be removed by polishing the specimen carefully and lightly with 0.05 micron alumina powder for 6 hours, which is relatively a very long operations compared to the conventional one.

From these facts it can be concluded that for γ phase the difference in cooling rare does not produce any structural differences. This fact is in agreement with the phase diagram. The hatchet marks, since they disappeared by prolong polishing, then they must be mechanical twins which form under slight plastic deformation during the sample grinding for the metallographic preparation. Associated with this twinning is a strong tendency toward intergranular cracking. These cracks appear to be the "wide grain boundaries" mentioned by Hume Rothery in the discussion of Murphy's paper(3).

3. *Widmanstatten Microstructure*

According to the phase diagram in Figure 1, both the 23.0 wt % Sn and the 20.0 wt % Sn alloys, at room temperature contain both β and γ phase. At homogenization temperature, which was 400°C, the 23.0 wt % Sn alloy contains also both phases, while the 20.0 wt % Sn alloy consist of γ phase only.

On fast cooling from the homogenization temperature, both alloys retain their high temperature microstructures. The 23.0 wt % Sn composes of β and γ phases as shown in Figure 4 and the 20.0 wt % Sn alloy consists of β phase only as shown in Figure 5. The dark phase of micrograph in Figure 4, which has a dendrite morphology, is the primary β phase. The lighter phase, which is the interdendritic fill, corresponding to the peritectic reaction of some of the primary with the liquid to form γ phase.

Figure 2. Microstructure of γ phase or Ag_3Sn
Furnace cooled from 400°C; mag. 400 X

Figure 3. Microstructure of γ phase or Ag_3Sn
Quenched in water from 400°C; mag. 400 X

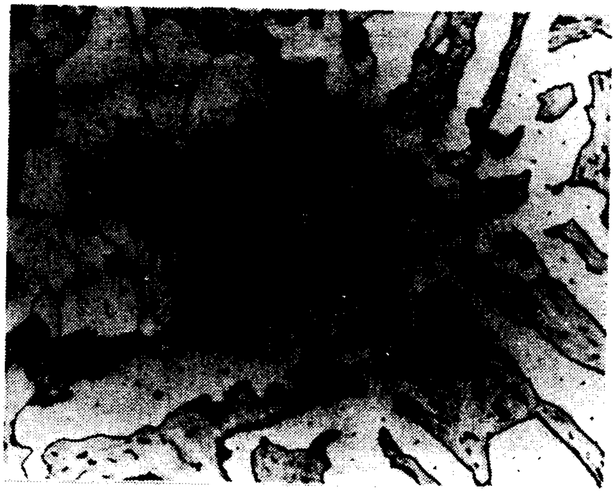

Figure 4. Microstructure of 23.0 wt % Sn alloy
Quenched in water from 400°C; mag. 400 X

Figure 5. Microstructure of 20.0 wt % Sn alloy
Quenched in water from 400°C; mag. 400 X

The phase diagram shows that there is no change of tin content in phase region will occur with the changing of temperature, but it shows that the tin solubility limit of β decreases with decreasing temperature. Therefore during slow cooling, β phase will reject its excess tin in the form of γ precipitates. This precipitation takes place on a certain crystallographic plane of β phase to form a "basket-weave pattern" or Widmanstatten structure as shown in Figure 6 for the 23.0 wt % Sn alloy and in Figure 7 for the 20.0 wt % Sn alloy.

The Widmanstatten structures in Figure 6 and 7 show as many as three trace directions in a single grain. These multiple markings could not correspond to the single basal plane of the HCP - β phase. The reject planes must therefore correspond to prism or pyramidal planes of the parent β phase. Presumably the β - HCP and γ - orthorhombic lattices have a particularly good match across the crystallographic interface plane. With this explanation Murphy's interpretation of the structure is varified except for his suggestion that the rejection is along cleavage plane of the matrix.

4. *Commercial Chip Alloys*

Based on the above explanation a conclusion can be drawn that commercial chip alloys which have higher in Ag (or alloy equivalent) than Ag_3Sn and have been slowly cooled from homogenization temperature will exhibit regions of Widmanstatten as shown in Figure 8. Plastic deformation during chipping probably involves movement of dislocations across these interfaces, β - γ, disorganizing the region of separation. As shown in Figure 9, the Widmanstatten pattern in chip alloy may be suppressed by fast cooling from the homogenization temperature. "Aging" of ingot chip alloy may be the rectification of unstable atomic configurations resulting from non-equilibrium cooling or plastic deformation.

Commercial alloys must represent some empirical balance between composition, homogenization schedule, plastic deformation and "Aging" to secure optimum manipulative and service characteristics in the amalgam restoration(5). Better understanding of the origin of structure should lead to systematic development of improved alloys.

CONCLUSION

1. "Tire tread" and "hatchet mark" patterns in homogenized

Figure 6. Microstructure of 23.0 wt % Sn alloy
Furnace cooled from 400°C; mag. 400 X

Figure 7. Microstructure of 20.0 wt % Sn alloy
Furnace cooled from 400°C; mag. 400 X

Figure 8. Microstructure of commercial alloy*)
Furnace cooled from 400°C; mag. 400 X

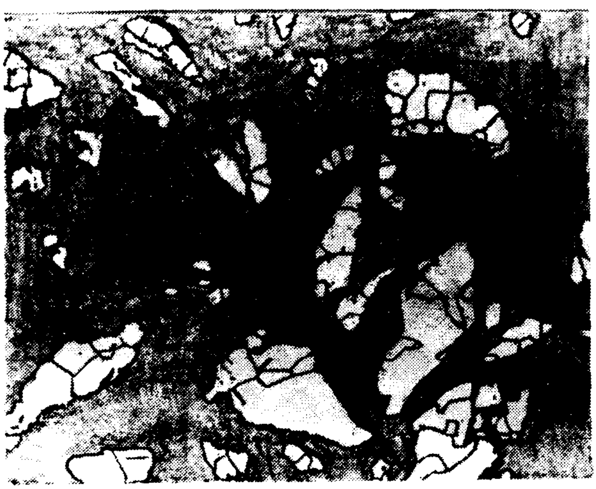

Figure 9. Microstructure of commercial alloy*)
Quenched in water from 400°C; mag. 400 X

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*) Traced from prints

γ - Ag_3Sn are deformation twins which result from plastic deformation in the early stages of metallographic prépara-tion. The "wide grain boundaries" also reported by Murphy[3] are intergranular cracks caused by mechanical stresses.

2. The microstructure of $\beta + \gamma$ alloys depends on cooling rate from the homogenization temperature. These differences should have their counterpart in manipulative and service behaviour of amalgams.

3. The Widmanstatten structure is formed during the slow cooling of β, which is supersaturated with γ, from the homogenizing temperature. During this slow cooling process γ is rejected and precipitates on favored crystallographic planes of the β parent lattice to generate the Widmanstatten structure.

4. Commercial chip alloys must have higher in alloy equivalent Ag than the stochiometric Ag_3Sn or γ, which make them capable of producing Widmanstatten structure.

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