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Amalgamation mechanism in dental amalgam alloys

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Model experiments to explore the amalgamation mechanism in conventional Ag_3Sn and high copper single-composition dental alloys have been conducted by rotating alloy rods in liquid mercury at different speeds and for different durations. After removing the rod from the mercury, the vacuum volatilization technique was used to accelerate the supersaturation of dissolved elements in the liquid mercury. The surface and sectional scanning electron microscope (SEM) views give direct evidence for new amalgamation mechanism in high copper dental alloys. The amalgamation reaction begins with the selective dissolution of the Ag_3Sn phase in mercury, tin gettering to the crumbled-off Cu_3Sn phase, heterogeneous nucleation of the Cu_6Sn_5 phase on the seed of the crumbled-off Cu_3Sn phase and was followed by the nucleation of the Ag_2Hg_3 phase. The unexplained phenomena, the formation of the γ-2 phase ($Sn_{7-8}Hg$) in higher content of mercury and the nonexistence of the γ-2 phase in higher content of copper alloys such as Sybralloy, were clearly understood by applying the new model.

1. Introduction

The addition of copper to the conventional Ag_3Sn dental amalgam has proved an effective way to remove the corrosive γ-2 ($Sn_{7-8}Hg$) phase from the final products of the amalgamation reaction. The copper-rich amalgams have showed superior marginal integrity and corrosion resistivity. There has been much research [1–10] on the metallography of hardened amalgam, the effects of processes such as trituration, condensation, mercury-alloy ratio, and the dimensional changes that occur during the hardening process to clarify the nature of this reaction. However the amalgamation reaction involves the liquid mercury at ambient temperature, and the alloys are composed of fine multi-phases. As a consequence, it is very difficult to investigate in detail how the amalgamation reaction takes place in real dental amalgam systems.

The copper is mostly present as Cu_3Sn intermetallic compounds in single-composition alloy powder or as additive particles of Ag–Cu eutectic in dispersal type alloy [9]. These single-composition alloy powders are manufactured by the atomizing process. They are mainly composed of the fine multi-phases such as ε (Cu_3Sn) and γ(Ag_3Sn) [11]. After they react with the liquid mercury, various reaction products are known to be formed by the dissolution–precipitation mechanism such as the conventional Ag_3Sn dental amalgam [2].

To explain the existence of the η-phase (Cu_6Sn_5) between original powders and silver amalgam (Ag_2Hg_3), the precipitation of the phase in advance of the silver amalgam phase in the liquid mercury phase was proposed by Okabe *et al.* [2–4]. They assumed that there are no differences in dissolution rate between silver, tin, and copper in liquid mercury. The solubilities in mercury at room temperature are 0.006 at % for copper, 0.066 at % for silver and 1.0 at % for tin. Since the dissolution rates are assumed to be the same, copper will reach its supersaturated concentration in mercury in advance of silver or tin. The supersaturated with copper should be relieved by the precipitation of the η crystals. Next to this precipitation, the mercury supersaturated with silver should be subsequently relieved by the precipitation of the Ag_2Hg_3 crystals.

But according to Hinzer and Stevenson [12], the dissolution rate differences in liquid mercury are large, in the following increasing order of copper, silver, tin. Thus the assumption of Okabe *et al.* was not appropriate to support their amalgamation reaction model. Another important consideration is that the dissolution behaviour of each phase in the powders is not the same. Because they are mainly composed of Ag_3Sn and Cu_3Sn phases, it should be understood which phase is more soluble in mercury and how they contribute to the amalgamation reaction. Another shortcoming of Okabe's model is that the occurrence of $Sn_{7-8}Hg$ phase in higher ratio of mercury can not be explained. To explain the formation of $Sn_{7-8}Hg$ phase in excess mercury content, Mahler [8] suggested another model, that the Cu_3Sn phase reacts with mercury faster than the Ag_3Sn phase. The tin liberated from the Ag_3Sn–Hg reaction reacts with the dissolved Cu_3Sn, forming the Cu_6Sn_5 phase. If there is excess mercury, the tin-gettering agents (Cu_3Sn) to form the Cu_6Sn_5 phase are not available. The dissolved excess tin should precipitate as the $Sn_{7-8}Hg$ phase to relieve the supersaturation.

Mahler's model can explain the existence of $Sn_{7-8}Hg$ phase in copper-rich amalgam but there is no

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ground for faster reaction of the Cu_3Sn phase with tin than that of the Ag_3Sn phase, and he does not give any direct metallographic evidence for his model.

The intent of this paper is to clarify how the high copper amalgam alloy reacts with mercury. A model experiment was conducted by rotating rods in a constant volume of liquid mercury (a simulation of trituration) and evaporating the remaining liquid mercury on the removed rod by exposure to the high vacuum atmosphere (10^{-5} torr). In real amalgam, the saturation of liquid phase with the dissolved elements takes place by the fast diffusion of mercury into the powders. The surface and the cross section of the rod were observed and analysed under a scanning electron microscope equipped with a microprobe analyser to study how this amalgamation reaction takes place.

2. Experimental procedure

Bars (5 mm diameter, 50 mm long) of conventional single phase Ag_3Sn(73.45 wt % Ag, 26.55 wt % Sn) alloy and high copper single-composition alloy (60 wt % Ag, 27 wt % Sn, and 13 wt % Cu) were prepared by centrifugal casting of the small melting stocks into a cast iron mould. The stocks were pre-alloyed in a vacuum induction melting furnace with a pure graphite crucible. The vacuum level for melting was 10^{-3} torr and purities of elements used in alloying were 99.9 wt %. Preheating of the mould was avoided and it was quenched into water immediately after casting. After annealing Ag_3Sn alloy in flowing argon at 703 K for 84 h and high copper alloy at 573 K for 4 h, the specimens were cut and examined by an X-ray diffractometer to confirm whether the bars have the same phases as in real dental amalgam.

To simulate the amalgamation reaction in dental amalgam, the cut and drilled specimens (5 mm diameter, 6 mm long) from the bars were rotated in a vessel containing mercury as in Fig. 1. The reaction vessel was maintained at 323 K in a silicon oil bath. To avoid the oxidation, argon gas was flowed over the liquid mercury. After rotating the specimen in the constant volume (3 ml) of liquid mercury at a different speed and time, the rotating rod was withdrawn from vessel. The remaining liquid on the specimen was evaporated with the aid of high vacuum. If the real amalgamation reaction proceeds with the dissolution and precipitation mechanism as proposed by many researchers, many reaction products should be observed on the surface of specimens.

Rotating the rod induces the dissolution of alloying elements in liquid mercury. Thus the rotating procedure corresponds to the triturating procedure in dental amalgam. The vacuum evaporating treatment gives the accelerated simulation of saturation with the dissolved elements in the liquid phase, which occurs in real amalgam due to the fast diffusion of mercury into the powders.

The reaction products on the surface were observed by a scanning electron microscope (ISI DS-130) and analysed by an energy dispersive spectrometer (EDX) (Philips PV9100). The take-off angle for the microanalysis was 65° and the Frame-C program was used to calculate the ZAF factors.

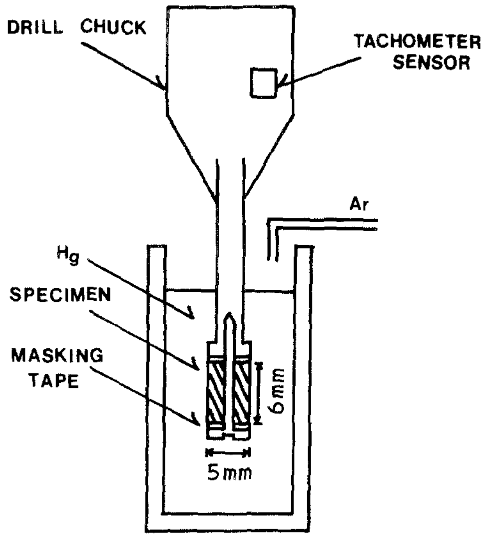


Figure 1 A schematic view of an apparatus for simulating a dental amalgamation reaction.

3. Results and discussion

3.1. Amalgamation mechanism of conventional Ag_3Sn alloy

Before studying the complicated high copper alloy composed of Ag_3Sn and Cu_3Sn phases, the Ag_3Sn alloy which has been used for a century as a standard dental amalgam was studied by this rotating rod technique. Amalgamation mechanism in this conventional Ag_3Sn alloy had not been clarified until Schoenfeld and Greener [13] showed that the amalgam reaction products nucleate from the liquid between the silver–tin particles and not from a solid solution within the particles. This result supports the idea that amalgamation involves dissolution of silver–tin powder in liquid mercury with the subsequent nucleation and growth of reaction products from the liquid mercury solution. Reynolds *et al.* [14] have shown direct evidence for the dissolution–precipitation mechanism by *in situ* scanning electron microscopy. They observed *in situ* that the reaction products nucleate and grow in liquid phase immediately after triturating the powder with mercury in a 1 : 1 ratio. They have characterized the amalgamation reaction in three stages, called the heavy alloy mechanism [15]:

1. particle rearrangement and the flow of liquid into empty spaces;
2. solution and precipitation, and
3. coalescence of grains and the merging of new phases with each other.

However, applying the heavy alloy mechanism to dental amalgam has several problems. In dental amalgam, the liquid phase is added by artificial means. Also, the liquid mercury is so transient that it disappears by interdiffusion while the compacts are at the sintering temperature, human body temperature. Another procedure unique to dental amalgam is the trituration process, which is the vigorous mixing step to start the reaction of the powder with mercury. However the role of this trituration step is not fully understood, and it should be clarified to study the mechanism of dental amalgam reaction. To simulate the trituration

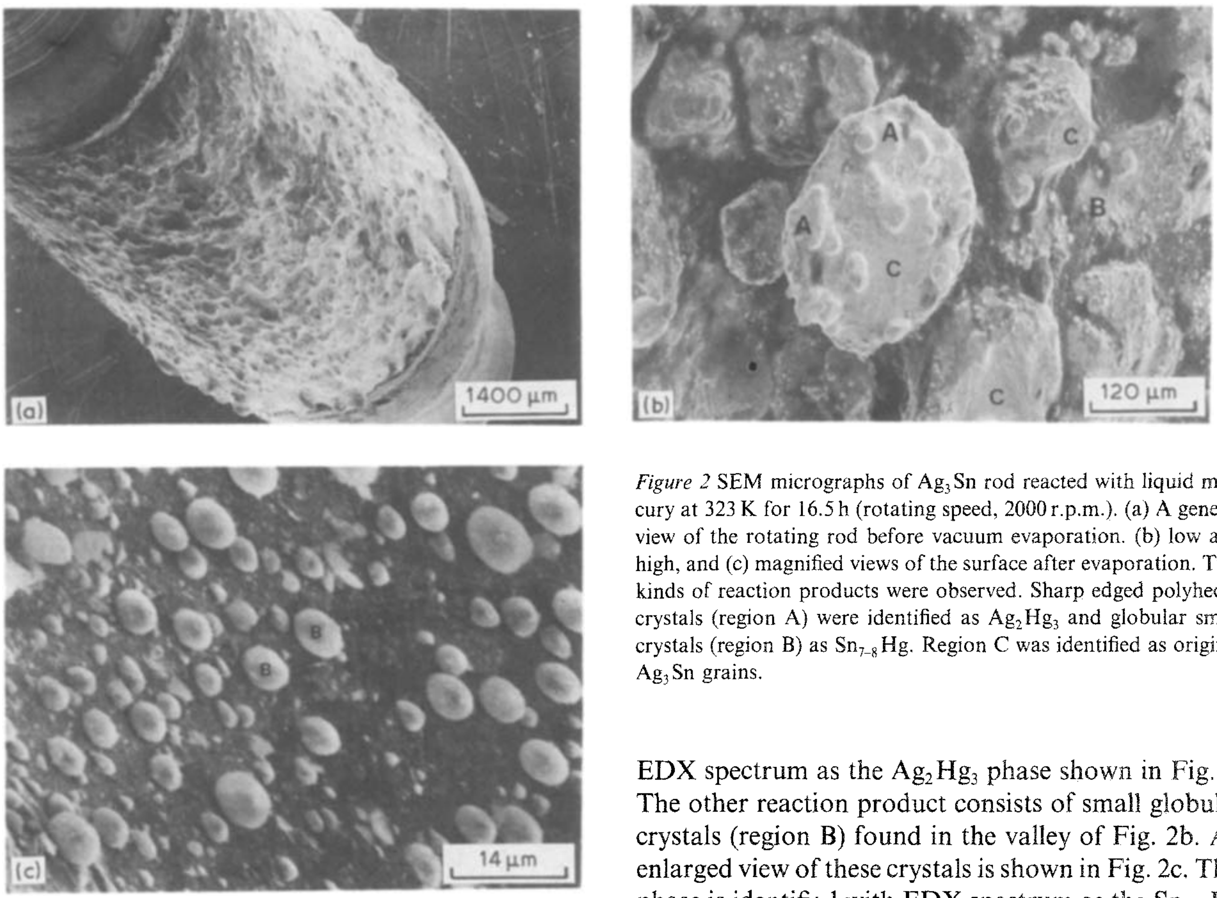
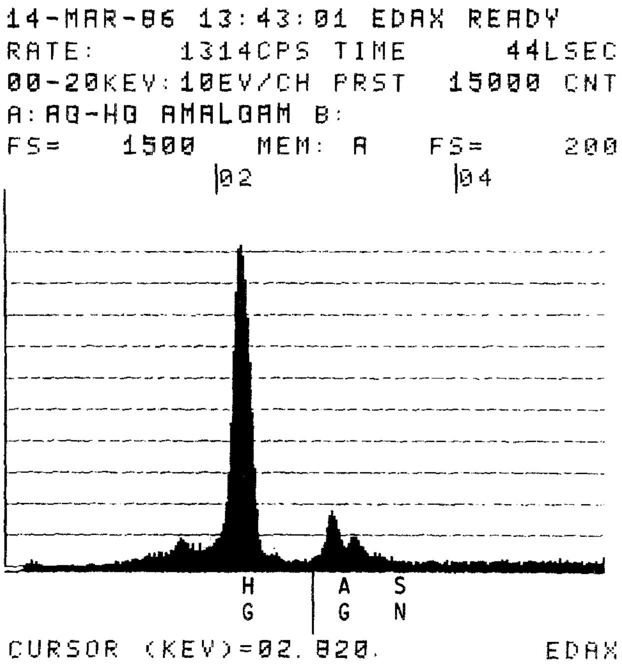


Figure 2 SEM micrographs of Ag_3Sn rod reacted with liquid mercury at 323 K for 16.5 h (rotating speed, 2000 r.p.m.). (a) A general view of the rotating rod before vacuum evaporation. (b) low and high, and (c) magnified views of the surface after evaporation. Two kinds of reaction products were observed. Sharp edged polyhedra crystals (region A) were identified as Ag_2Hg_3 and globular small crystals (region B) as $Sn_{7-8}Hg$. Region C was identified as original Ag_3Sn grains.

process, the Ag_3Sn rod is rotated in liquid mercury with different speeds and time periods.

Fig. 2a shows a general view of the rotating rod after rotating at 2000 r.p.m. for 16.5 h. Liquid mercury is observed on the surface of the rotating rod. After evaporating the liquid mercury in a high vacuum chamber for 5 h, the surface of this rod changed to that seen in Fig. 2b and c. In Fig. 2b, two kinds of reaction products are observed. One is sharp edged small polyhedra crystals (region A) identified with EDX spectrum as the Ag_2Hg_3 phase shown in Fig. 3. The other reaction product consists of small globular crystals (region B) found in the valley of Fig. 2b. An enlarged view of these crystals is shown in Fig. 2c. This phase is identified with EDX spectrum as the $Sn_{7-8}Hg$ phase shown in Fig. 4. The bulky crystals (region C) in the centre of Fig. 2b are identified as the original Ag_3Sn phase.

If reaction products are formed from the saturated solution of mercury, changes in dissolution conditions should give any difference in the quantity of the reaction products. Fig. 5a shows the surface of the rod which suffered least severe dissolution (250 r.p.m., 20 min). Almost no reaction products are seen in this photograph. The reason is that the quantities of silver and tin dissolved in liquid phase are not enough to



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00-20KEV:10EV/CH PRST  15000 CNT
A:AG-HG AMALGAM B:
FS=   1500    MEM: A    FS=    200
           |02            |04
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CURSOR (KEV)=02.820.                EDAX
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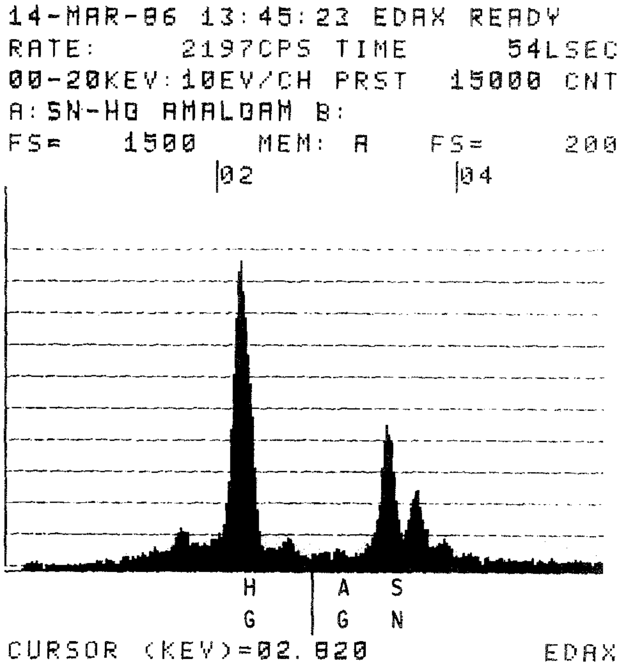
Figure 3 Typical energy dispersive spectrum for the sharp edged polyhedra crystals (region A in Fig. 2b) identified as γ_1-phase (Ag_2Hg_3).



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00-20KEV:10EV/CH PRST  15000 CNT
A:SN-HG AMALGAM B:
FS=   1500    MEM: A    FS=    200
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CURSOR (KEV)=02.820                 EDAX
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Figure 4 Typical energy dispersive spectrum for the small globular crystals (region B in Fig. 2b) identified as γ_2-phase ($Sn_{7-8}Hg$).

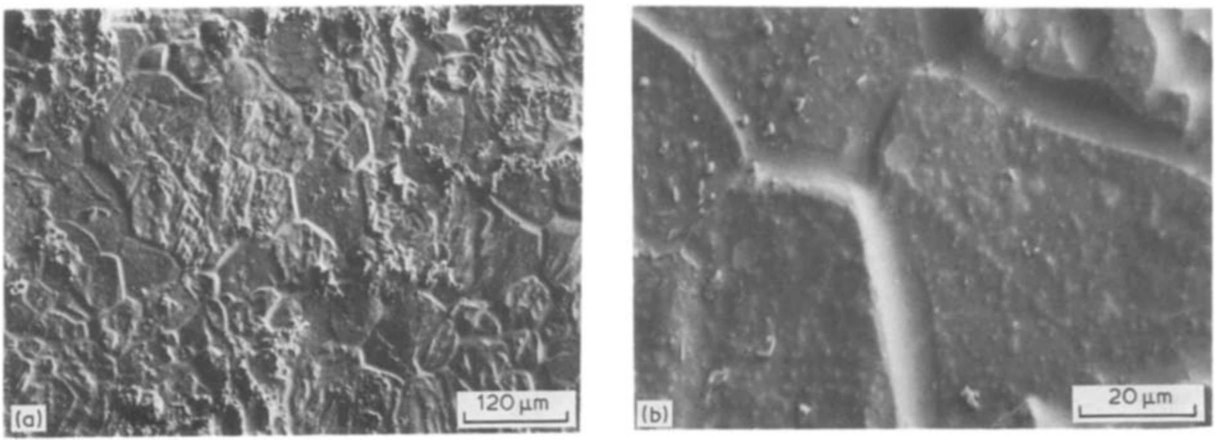


Figure 5 Enlarged surface views of Ag_3Sn rod reacted with mercury at 323 K for 20 min (rotating speed, 250 r.p.m.). (a) low and (b) high magnified micrographs showing that almost no reaction products were precipitated due to the insufficient dissolution of Ag_3Sn phase in liquid phase.

precipitate Ag_2Hg_3 and $Sn_{7-8}Hg$ crystals. Instead of finding reaction products, sharp grain boundaries of the original Ag_3Sn phase are well delineated by the selective mercury attack on the grain boundary. The enlarged view (Fig. 5b) also shows the absence of reaction products and the well developed grain boundaries of the Ag_3Sn phase without using any special etching agent.

If more severe dissolution conditions are applied to the specimen, precipitated crystals should be observed on the surface of the rod. Fig. 6 shows the surface view of the rotating rod which suffered more severe dissolution (2000 r.p.m., 2 h). The irregular surface shows that the rod is attacked more severely by the liquid mercury than the one in Fig. 5. In Fig. 6b, various reaction products are observed. The long rod polyhedra crystals are identified as Ag_2Hg_3, and the small globular crystals, the $Sn_{7-8}Hg$ phase. It is observed that there is a remaining liquid mercury film in the circled region A in Fig. 6b. Also the circled region B shows the Ag_2Hg_3 crystals which have grown vertically.

These findings are direct evidence that reaction products in conventional dental amalgam are formed in liquid phase by the dissolution–precipitation mechanism. From the above observed results, it can be concluded that the amalgamation reaction of Ag_3Sn powder has two major reaction steps: the dissolution of Ag_3Sn powder and the precipitation of Ag_2Hg_3 and $Sn_{7-8}Hg$ crystals. In the dissolution step, three silver atoms and ore tin atom are liberated from the alloy powders per single step of the dissolution. The concentration of the dissolved silver should reach its saturated value faster than that of tin because of its larger amount of the dissolvable quantity and lower solubility in mercury. This is the reason why the Ag_2Hg_3 crystals are formed faster than the $Sn_{7-8}Hg$ crystals and the reaction products of conventional alloys are mainly composed of the Ag_2Hg_3 phase [16].

This saturation is promoted by the fast mercury diffusion into alloy powders. Mercury can diffuse very rapidly into the Ag_3Sn alloy through the grain boundaries. The order of grain boundary diffusivity is known to be $10^{-3}\,cm^2\,sec^{-1}$ at 323 K [17].

The supersaturated liquid phase with silver should be relieved by the precipitation of Ag_2Hg_3 crystals. Next to that precipitation, the supersaturation with tin is relieved by the precipitation of $Sn_{7-8}Hg$ phase.

3.2. Amalgamation mechanism of high copper single-composition alloy

After Asgar's finding [1] that high copper single-composition alloy powders showed the same behaviour as high copper dispersal alloy powders, various high copper alloy powders have been introduced into the market. These alloy powders have the range of 13 to

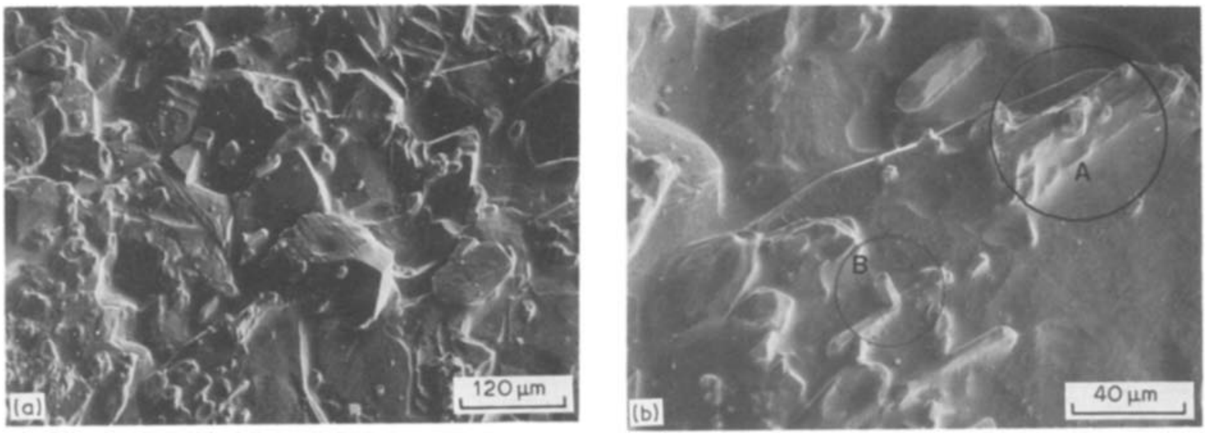


Figure 6 Enlarged surface views of Ag_3Sn rod reacted with mercury at 323 K for 2 h (rotating speed, 2000 r.p.m.). More severe dissolution made the surface of the original Ag_3Sn rod irregular. Observed long rod and small polyhedra crystals were all identified as Ag_2Hg_3 phase.

TABLE I Results of X-ray diffractometry and data for each phase (2θ and I/I_0 values)

Real alloy powders	As casted rod	Heat-treated rod	Ag_3Sn (γ)	Ag–Sn (β)	Ag–Sn eutectic	Cu_3Sn (ε)
34.7(27)	34.7(34)	34.7(25)	34.64(60)	35.34(12)	30.84(40)	–
					32.08(100)	
37.6(46)	37.6(43)	37.6(51)	37.64(80)	37.64(100)		–
39.7(100)	39.6(100)	39.7(100)	39.52(100)	–	–	–
41.5(1)	41.3(16)	41.7(10)	–	–	–	41.8(40)
43.2(21)	43.0(25)	43.2(27)	–	–	44.86(50)	43.5(100)
52.2(20)	52.3(21)	52.3(21)	51.96(80)	52.6(12)	–	–
–	57.1(14)	57.4(7)	–	–	–	57.6(27)
62.4(15)	62.3(19)	62.4(17)	61.86(80)	63.26(12)	64.74(20)	–
69.3(20)	69.2(19)	69.3(25)	69.06(80)	69.64(19)	–	67.92(30)
			72.74(50)	75.44(19)	72.74(15)	–
75.1(16)	75.0(16)	75.2(19)	76.16(90)			76.88(40)

30 wt % Cu content, and are composed of fine phases due to rapid cooling in the atomizing process. According to Kraft and Petzow [11], high copper single composition alloy powers were mainly composed of Ag_3Sn and Cu_3Sn phases. To discover the role of these phases on the amalgamation reaction, the same model experiment as in section 3.1 was carried out. To obtain a similar cooling rate as for real alloy powders, preheating of the mould was avoided, and the mould was water-quenched immediately after centrifugal casting. The results of X-ray diffraction of the commercial amalgam alloy powder (Tytin, S. S. White Dental Products International, Philadelphia, USA) and the specimen for model experiments are given in Table I. The specimens have the same structure as commercial dental amalgam alloy, but the constituent phases are larger than for the commercial alloy powder due to their slower cooling rate. After this specimen reacting with liquid mercury in various conditions, the surface of the rod was observed as in Fig. 7. The rotating condition for Fig. 7a was 2000 r.p.m. for 2 h, and that of Fig. 7b was 250 r.p.m. for 20 min. Two types of reaction products are observed in both specimens. The long rod and the small disc crystals (region B) were identified as η-phase(Cu_6Sn_5) and large polyhedra crystals (region A) are γ-1 phase(Ag_2Hg_3) by EDX. The higher r.p.m. can give more dissolution of the rotating rod, resulting in larger reaction products. The sectional view of these specimens is shown in Fig. 8b and c. For a comparison, Ag_3Sn rod which was subjected to the dissolution condition at 2000 r.p.m. for 16.5 h is also presented in Fig. 8a. This photograph was taken as a polished state. A sharp grain boundary is well delineated due to the mercury attack into the grain boundary of Ag_3Sn phase. The sectional view of high copper alloy in Fig. 8b and c shows the reaction products and the unreacted region of the rotating rod. The unreacted original alloy shows a two phase structure. The grey phases in the circled region in Fig. 8b and c are identified by the point analysis of EDX as Cu_3Sn and the white ones as Ag_3Sn. At the top of Fig. 8b, the reaction products are observed as a group of smaller separated crystals. Unreacted rod is observed at the bottom of the photograph which shows the behaviour of each phase in amalgamation reactions. These micrographs give important metallurgical evidence for the amalgamation mechanism. As shown in the circled region, the grey Cu_3Sn phase is observed while the white Ag_3Sn phase is not. The crumbling-off phenomenon of the grey Cu_3Sn phases is also shown. These facts show that the Ag_3Sn phase is selectively dissolved into the liquid mercury while the Cu_3Sn phase is not dissolved in liquid mercury. In the process of the selective dissolution of the Ag_3Sn phase, the Cu_3Sn is crumbled-off into the liquid mercury as the Cu_3Sn state. The Cu_3Sn phase collects the dissolved tin to form the more stable Cu_6Sn_5 phase. These metallographic observations give direct evidence that the amalgamation reaction does not take place by the mechanism proposed by Okabe *et al.* [2–4]. A schematic drawing of their model is given in Fig. 9. As discussed in the introduction, this model has a mistake

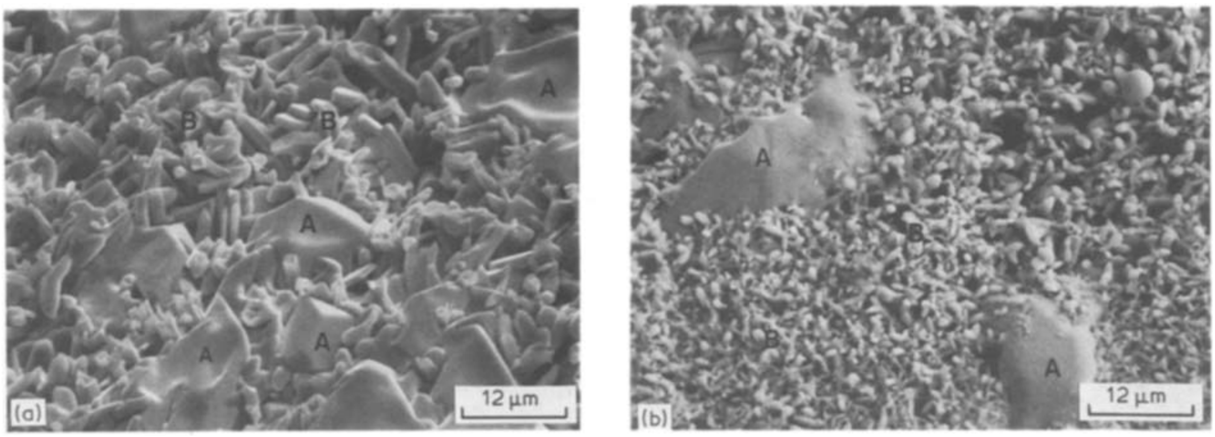
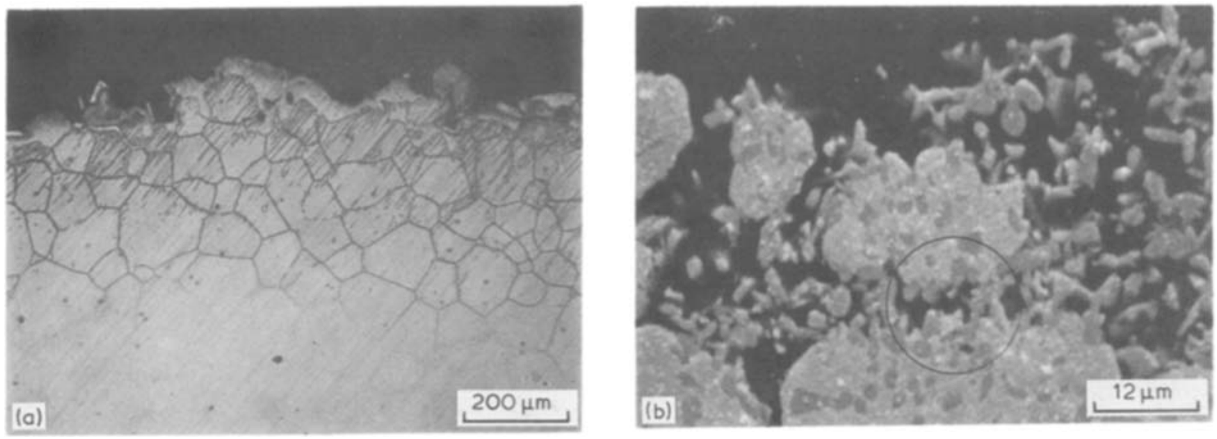


Figure 7 Enlarged surface views of high copper alloy after rotating in different conditions. Long rod and small disc-type crystals (region B) were identified as η-phase (Cu_6Sn_5) and bigger polyhedra crystals (region A) were identified as γ_1-phase (Ag_2Hg_3). More severe rotation made the bar dissolve further, and resulted in larger reaction products. (a) 2000 r.p.m., 2 h; (b) 250 r.p.m., 20 min.



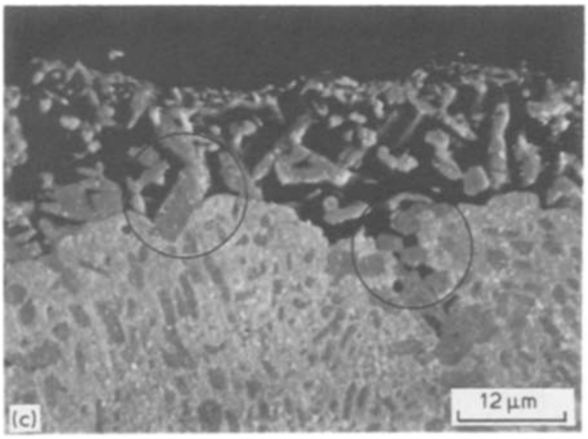


Figure 8 Sectional views of rotating rods after rotating under different conditions. (a) Ag_3Sn rod, 2000 r.p.m., 16.5 h: optical Micrograph. (b) High copper alloy, 2000 r.p.m., 2 h: SEM micrograph. (c) High copper alloy, 250 r.p.m., 20 min: SEM micrograph.

in its basic assumption. Forming the $Sn_{7-8}Hg$ phase in a high ratio of mercury cannot be explained. But a new amalgamation mechanism based on the findings of this research can clarify the unexplained observations in high copper dental amalgam. The proposed reaction steps are summarized as follows:

1. Selective dissolution of the Ag_3Sn phase into mercury during trituration.
2. Crumbling-off of the Cu_3Sn phase into the liquid phase.
3. Supersaturation of the liquid phase due to mercury diffusion into the alloy powders.
4. Tin gettering to the Cu_3Sn phase and heterogeneous nucleation and growth of the Cu_6Sn_5 phase on the Cu_3Sn seed.
5. Nucleation and growth of the Ag_2Hg_3 phase.

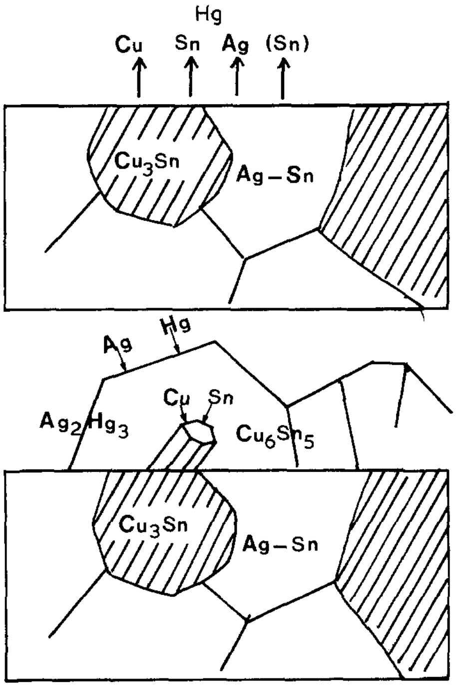


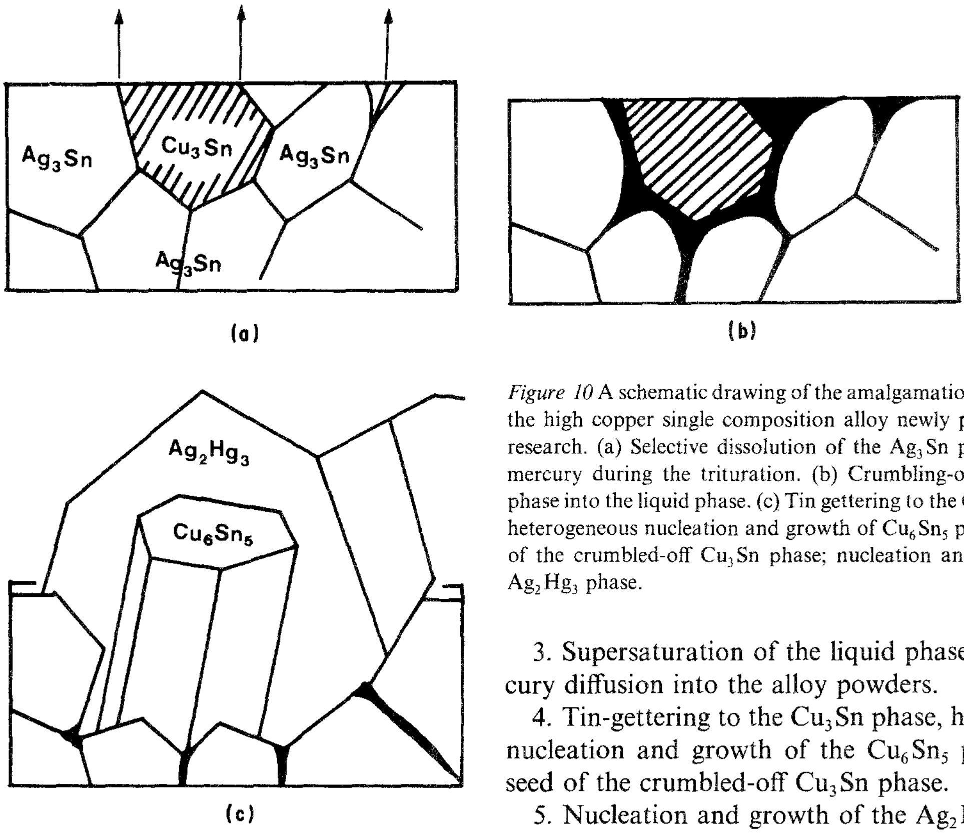
Figure 9 A schematic drawing of the proposed amalgamation mechanism for high copper single composition alloy by Okabe *et al.* [2–4].

A schematic drawing of the mechanism proposed by this research is given in Fig. 10. To explain the formation of the corrosible γ-2 phase($Sn_{7-8}Hg$) in a high copper spherical alloy such as Tytin, a four-phase equilibrium theory is suggested by Kraft and Petzow [11]. However this theory is unable to explain why γ-2 phase does not exist in lower content of mercury [8]. Applying the mechanism proposed above, one can explain the reason for the formation of γ-2 phase in cases of excess mercury content. When excess mercury is added to the powders, the available tin gettering agents of the crumbled-off Cu_3Sn after trituration are limited in number because of continuous mercury attack on the Ag_3Sn phase and liberation of tin from this attack. To relieve the supersaturation of tin in the liquid phase, the tin amalgam phase($Sn_{7-8}Hg$) must nucleate and grow.

Another fact [11] that could not be explained by Okabe's model is that the higher copper amalgam alloys such as Sybralloy (Kerr Manufacturing Co., Michigan, USA) containing 30 wt % Cu do not show the corrosible γ-2 phase compared to the standard high copper alloy such as Tytin. This can also be explained by this proposed mechanism. If the copper content of the alloy system is increased, the Cu_3Sn phase available to catch the dissolved tin in liquid phase is also increased in number. So the tin is unable to reach the saturated concentration needed to precipitate the γ-2 phase.

4. Conclusions

The amalgamation mechanism of conventional Ag_3Sn alloys involves the dissolution of Ag_3Sn powder in the liquid mercury with subsequent nucleation and growth of reaction products from the liquid phase. If more severe dissolution conditions are applied to the



(a)

(b)

(c)

Figure 10 A schematic drawing of the amalgamation mechanism for the high copper single composition alloy newly proposed by this research. (a) Selective dissolution of the Ag_3Sn phase into liquid mercury during the trituration. (b) Crumbling-off of the Cu_3Sn phase into the liquid phase. (c) Tin gettering to the Cu_3Sn phase and heterogeneous nucleation and growth of Cu_6Sn_5 phase on the seed of the crumbled-off Cu_3Sn phase; nucleation and growth of the Ag_2Hg_3 phase.

alloy, more precipitates are formed in the liquid phase of mercury. Simple contact of the alloy with mercury does not produce any precipitates. Dissolution of the alloy occurs selectively along the grain boundary of the alloy. The alloy is dissolved as a Ag_3Sn intermetallic form. Three silver atoms and one tin atom are dissolved in liquid mercury per single dissolution step. This is the reason why reaction products are mainly composed of silver amalgam, Ag_2Hg_3 crystals. Since the solubility of silver in liquid mercury is lower than that of tin, and the dissolvable quantity of silver is three times that of tin, the Ag_2Hg_3 crystals are formed faster than $Sn_{7-8}Hg$ crystals to relieve the faster saturation.

In high copper single-composition alloys, the new amalgamation model based on experimental observation can explain the formation of the γ-2 phase in higher mercury content, and the nonexistence of the γ-2 phase in higher copper content alloys such as Sybralloy. The tin-gettering action of the crumbling-off Cu_3Sn phases has a most important role in prohibiting the formation of the corrosive γ-2 phase. The proposed reaction mechanism can be summarized as follows:

1. Selective dissolution of the Ag_3Sn phase into mercury during the trituration.
2. Crumbling-off of the Cu_3Sn phase into the liquid phase.
3. Supersaturation of the liquid phase due to mercury diffusion into the alloy powders.
4. Tin-gettering to the Cu_3Sn phase, heterogeneous nucleation and growth of the Cu_6Sn_5 phase on the seed of the crumbled-off Cu_3Sn phase.
5. Nucleation and growth of the Ag_2Hg_3 phase.

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