

STRUCTURE AND HARDNESS OF FOUR TYPES OF AMALGAM

BY

Toshio TAKATSU, Tadao IKEDA, Chioko HIGASHI,
Masaaki IWAKU and Takao FUSAYAMA*¹

ABSTRACT

Three non- γ_2 and one conventional amalgams were compared for their structure and hardness. The Ag-Cu dispersants hardened the amalgam by generating hard Cu-Sn halos and diminishing the γ_2 phase. The Ag-Sn-Cu-In single composition alloy produced the hardest amalgam by its high hardness and the Cu-Sn reaction phase scattered as minute granules.

INTRODUCTION

The authors previously investigated the metallurgical structure of a dispersion-strengthened amalgam*² and its clinical significance and presumed that the decreased marginal failures with this amalgam was mainly based on the chemical resistance improved by the absence of the γ_2 phase¹⁾.

This amalgam has shown remarkably better clinical results than the conventional amalgams²⁻⁴⁾ but some disadvantages have also been pointed out as follows: (1) the early strength is low, (2) the mercury-alloy ratio is critical for the optimum plasticity⁵⁾ and (3) accurate proportioning of the alloy powder is difficult with a dispenser. These disadvantages were considered to be mainly due to the fact that its powder is a mixture of lathe-cut alloy particles and spherical dispersant particles. Further investigation was carried out by Asger *et al.* to produce all spherical alloy powder of strengthened

type^{6,7)}. Recently a Japanese manufacture developed an alloy composed of γ and dispersant particles, both in spherical form, and then another alloy composed of spherical particles of single composition in which the γ particles were modified by adding copper and indium.

In this study, the metallurgical structure of the amalgams made from these new alloys was investigated by using optical microscopy, X-ray probe microanalysis and microhardness test.

MATERIALS AND METHODS

Four types of amalgam alloys (Table 1, Fig. 1) were proportioned with mercury according to the directions of the manufacturers and triturated with a mechanical amalgamator*³ for 15 seconds. The mixture was packed in a vinyl tube of 4 mm in inner diameter and 4 mm in height placed on a stainless steel matrix band with a hand condenser with a 3-mm round tip under a pres-

*¹ 高津寿夫, 池田忠雄, 東千緒子, 岩久正明, 総山孝雄: Department of Operative Dentistry (Chief: Prof. T. FUSAYAMA), School of Dentistry, Tokyo Medical and Dental University (Tokyo Ika Shika Daigaku).

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*² Dispersalloy, Code 2830, Johnson & Johnson Co. East Windsor, N.J., U.S.A.

*³ Shofu Amalgam Mixer Deluxe, Shofu Dental Mfg. Co., Ltd., Kyoto, Japan.

Table 1. Amalgam Alloys Used

Alloys (abbreviation)	Kind of particles	Form	Batch No.	Hg: alloy ratio	Pestle
Shofu Spherical ¹ (SS)	Ag-Sn spherical	Powder	331	0.85	Without
Dispersalloy ² (DA)	Ag-Sn lathe-cut Ag-Cu spherical	Pellet	2830	1.00	With
Spherical-D ¹ (SD)	Ag-Sn spherical Ag-Cu-Sn spherical	Powder	003	0.90	Without
Indiloy ¹ (IL)	Ag-Cu-Sn-In spherical	Powder	001	0.86	Without

1. Shofu Dental Mfg. Co., Ltd., Kyoto, Japan

2. Johnson & Johnson Co., East Windsor, N.J., U.S.A.

sure adjusted to 40 kg/cm² by a hanging type pressure regulator.⁸⁾ The filling was em-

bedded in a polyester resin at 24 hours after packing. After more than 6 days, the sur-

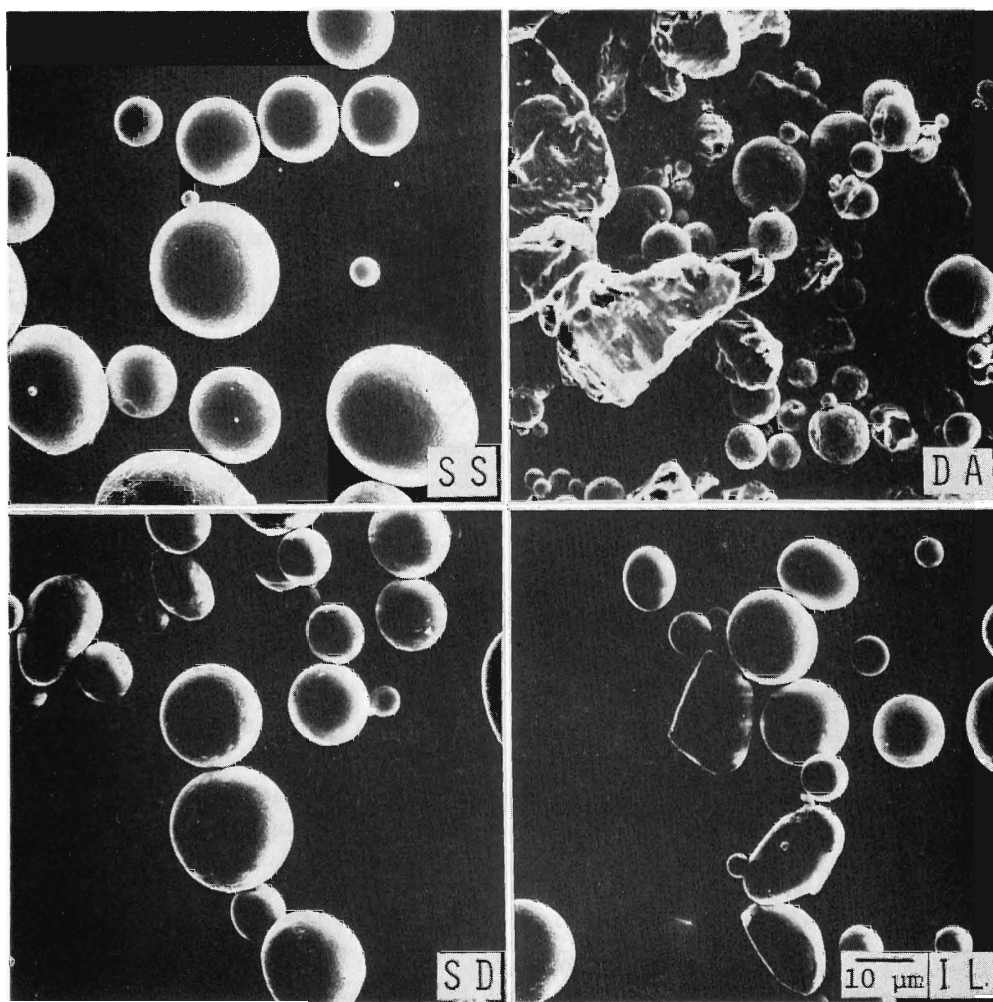


Fig. 1

face of the specimen set against the matrix band was reduced with emery papers under running water and polished with diamond pastes.

The polished surface was first observed under a metallurgical microscope. For observing the distribution of the metallic elements, the specimen surface was covered with vacuum-evaporated carbon, and the back-scattered electron images and the characteristic X-ray images of Ag, Cu, Sn, Hg, Zn, and In on the surface were observed at a magnification of $1,000\times$ by an electron probe microanalyzer.*⁴

The hardness of the individual phases of the amalgam was measured by pressing the Vickers indenter of Hanemann's microhard-

ness tester on the surface of more than 3 weeks of age with a 2.0-gram load for 20 seconds. The overall hardness of the amalgam surface was also measured with the Knoop indenter of the microhardness tester. Measurement was repeated 10 times for each phase.

RESULTS

Optical microscopic structure of the amalgams

When observed under an optical microscope (Fig. 2), the SS amalgam showed many rod-shaped or irregularly shaped γ_2 phases scattered in the matrix between the residual spherical γ particles.

The DA amalgam showed irregularly

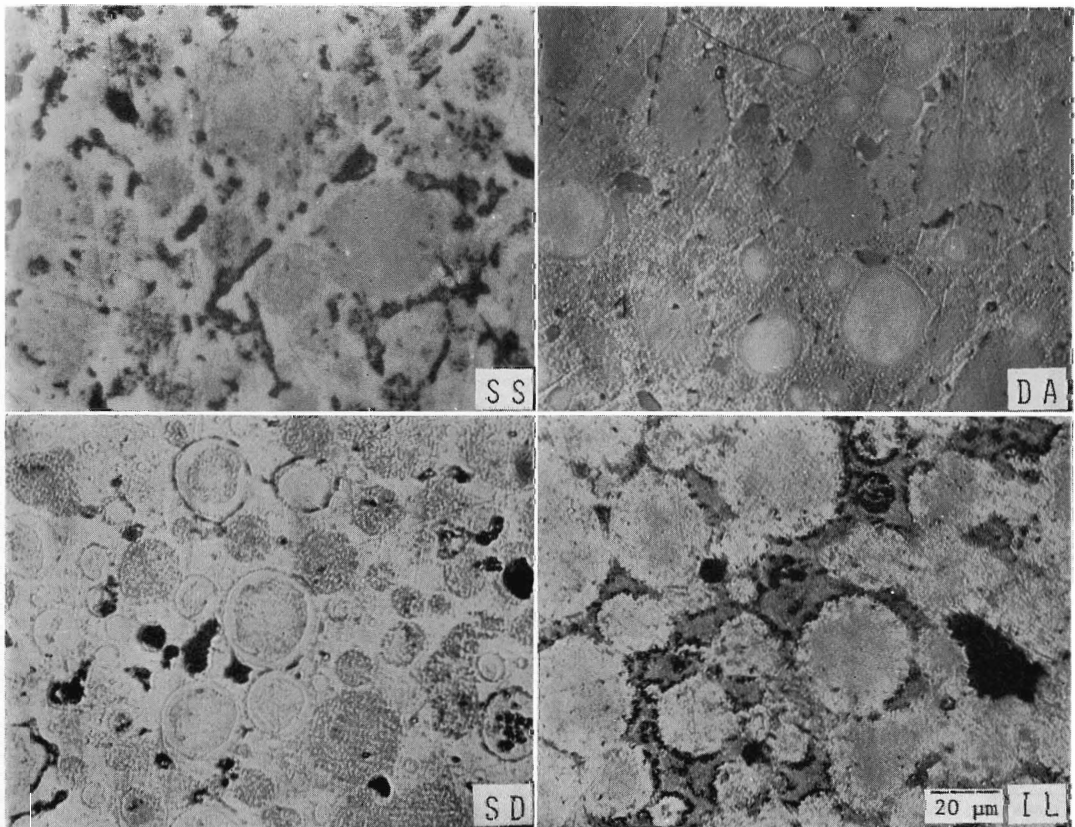


Fig. 2

*⁴ JSM-U3, Japan Electron Optics Laboratory, Tokyo, Japan.

shaped residual γ particles and spherical residual dispersant particles surrounded by halos. The γ_2 phase was hardly observed in the matrix. A few dark grayish blue minute particles (Cu-Sn, ϵ phase) were scattered in both the γ particles and the matrix. Minute cleft-like structures approximately $1\ \mu\text{m}$ in width were frequently observed in a chain form adjacent to the residual γ particles.

The SD amalgam showed spherical residual γ particles and spherical residual dispersant particles surrounded by halos. Tiny γ_2 phases were observed rarely in some parts of the matrix. A few voids were also observed in the matrix. Minute slender cleft-like structures were sometimes observed around the halo surrounding the residual dispersant particles.

The IL amalgam showed spherical residual alloy particles of uniform appearance surrounded by somewhat brighter halos, with irregular widths. No γ_2 phase was observed in the matrix.

Elemental composition of individual phases of the amalgams

When observed by an X-ray probe micro-

analyzer (Fig. 3), in the SS amalgam, the residual alloy particles and the matrix consisted respectively of Ag and Sn (γ) and of Ag and Hg (γ_1). Many rod or irregular-shaped structures consisting of Sn and Hg (γ_2) were observed in the matrix.

The DA amalgam had an elemental composition similar to the SS amalgam in regard to the residual alloy particles and the matrix. The residual dispersant particles consisted of Ag and Cu. The halos surrounding them consisted of Cu, Sn and small amounts of Ag and Hg. No structure consisting of Sn and Hg (γ_2) was observed. The elemental composition of the SD amalgam was generally similar to the DA amalgam, except for the small amount of Sn contained in the residual dispersant particles.

In the IL amalgam, the residual alloy particles consisted of Ag, Sn, Cu and a small amount of In, though the composition of the matrix was similar to that of the above amalgams. The halos surrounding the particles, in which new granular phases were scattered, contained not only Cu and Sn but also Ag and Hg.

Table 2. Hardness of Four Amalgams

Hardness measured	Alloy (type)	SS (conventional)	DA (dispersant-added)	SD (dispersant-added)	IL (single composition)
Hardness of individual phases (Vickers H. N.)	Ag-Hg+Sn-Hg matrix	109±17	—	—	—
	Ag-Hg matrix	—	X 143±11	140±14	151±17
	Ag-Sn residual alloy particles	153±23	159±18	174±11	—
	Ag-Cu-Sn-In residual alloy particles	—	—	—	X 247±17
	Ag-Cu residual dispersant particles	—	157±11	—	—
	Ag-Cu-Sn residual dispersant particles	—	—	167±24	—
Overall hardness (Knoop H. N.)	Cu-Sn-halos surrounding the dispersant particles	—	255±22	267±29	—
	Ag-Cu-Sn-In halos surrounding the alloy particles	—	—	—	X 226±17
Overall hardness (Knoop H. N.)		109±4	X 130±8	134±12	X 184±11

X indicates that the difference between the means of the left and right groups was statistically significant at the level of 95% confidence by the *t* test.

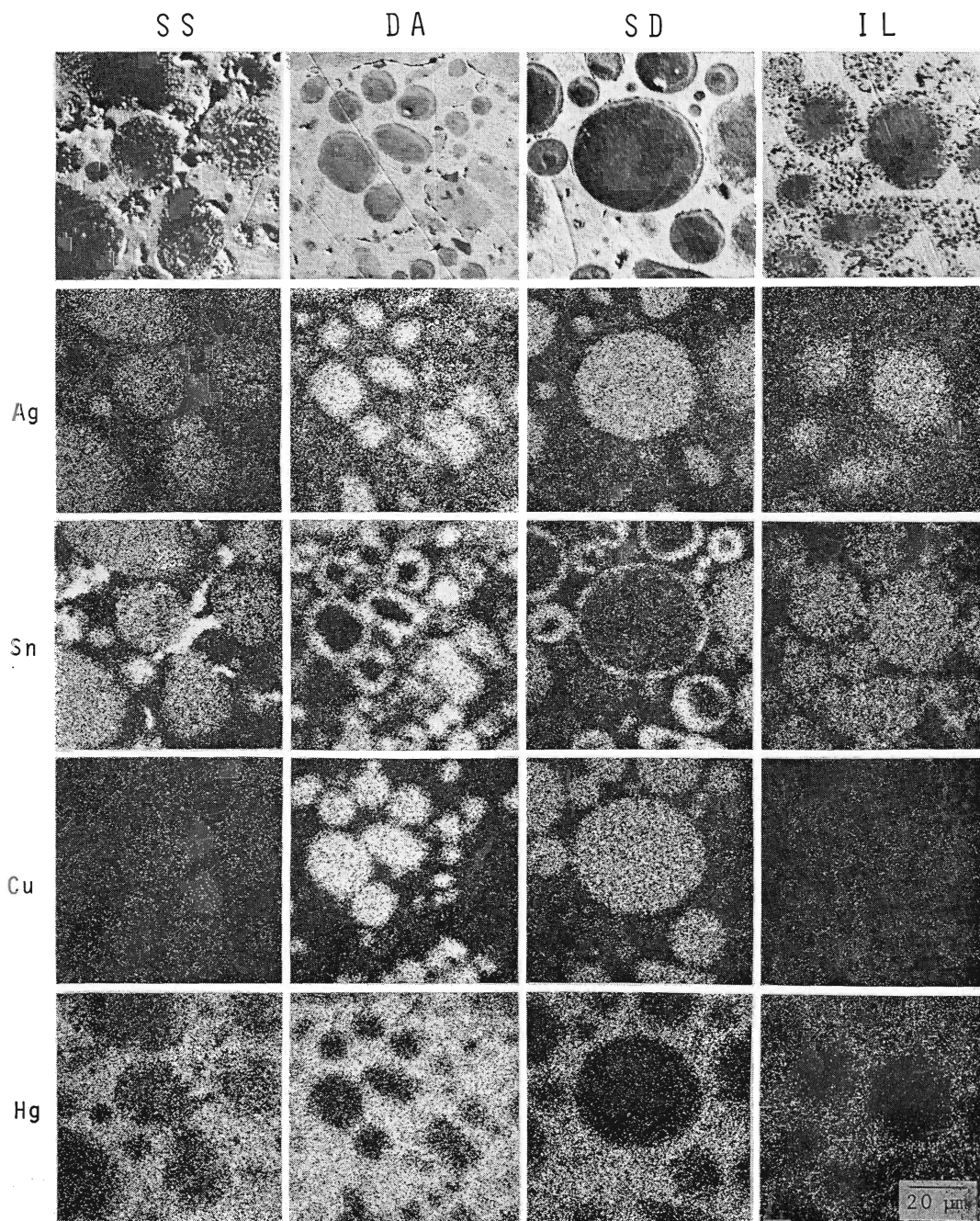


Fig. 3

Hardness of the amalgams

The hardness of the individual phases of the amalgams was compared as follows (Table 2): The hardness of the matrix was comparable in the DA, SD and IL amalgams but much lower in the SS amalgam. The hardness of the residual alloy particles was comparable in the SS, DA and SD amalgams but much higher in the IL amalgam. The hardness of the residual dispersant particles was comparable in the DA and SD amalgams and also to that of the residual alloy particles in the same amalgams. The hardness of the halos surrounding the dispersant particles in the DA and SD amalgams was higher than that of any of the matrices and residual alloys and dispersant particles and comparable in both amalgams. In the IL amalgam, the hardness of the halos was slightly lower than that of the residual alloy particles surrounded by the halos but much higher than that of the matrix. As compared with the hardness of the SS amalgam, the overall hardness of the DA and SD amalgams was 20% higher and that of the IL amalgam was 70% higher.

DISCUSSION

Alloy particles

The SD amalgam alloy with dispersant particles added and the IL amalgam alloy of single composition particles were both composed of only the spherical particles. The fact that they do not contain lathe-cut particles is considered to make accurate proportioning with a dispenser possible, decrease the required amount of mercury and thus simplify their use. Such amalgams made from the spherical particles are assumed to adapt more readily to the cavity walls⁹⁾.

The manufacturer of the SD amalgam reported that they encountered difficulty in producing small spherical dispersant particles, and that they were afraid of the possi-

ble separation of the two types of spherical particles of different specific gravities during storage, and so they turned to the IL amalgam alloy of single composition spherical particles, in which In was added to prevent tarnish or corrosion.

γ_2 phase

The γ_2 phase was hardly observed in the DA amalgam, in agreement with the previous reports^{1,10-12)}, and also in the newly developed SD and IL amalgams. The Sn from the γ particle (Ag-Sn) seemed to be all consumed in forming the Cu-Sn phase in the reaction with Cu, thus preventing the formation of the Sn-Hg phase (γ_2).

Hardness

The fact that the matrix was much harder in the three new types of amalgams than in the SS amalgam seems to be based on the absence of the soft γ_2 phase¹³⁾. This seems to be the first reason for the improved physical properties of the new amalgams. The second reason seems to be the presence of the halo-shaped reaction phases. The authors previously reported that the halo was much harder than the residual dispersant particles and was responsible for the improvement of the physical properties of the DA amalgam¹⁾. This was also found to be true with the SD amalgam in this experiment. The halo was slightly less hard in the IL amalgam, probably because it was composed of minute segments of the hard Cu-Sn phase scattered in the γ_1 phase. Such dispersants scattered in minute size are considered to be most effective for strengthening the amalgam.

The hardness of the residual alloy particles was also the highest in the IL amalgam. Such a high hardness of the residual alloy particles and the effective dispersion-strengthening seemed to produce an extremely high overall hardness in the IL amalgam.

Conjunction between the matrix and the residual particles

Mahler *et al.*¹²⁾ previously reported that the characteristic minute porosities were observed between the γ_1 matrix and the Cu-Sn halos surrounding the residual dispersant particles in the DA amalgam. In this study, similar minute porosities were observed also around the residual γ alloy particles of the DA amalgam and around the halos of the SD amalgam. The presence of such a structure indicating insufficient conjunction between the matrix and the particles is considered to be the factor affecting the strength of amalgam. The IL amalgam did not have such porosities. This may also be responsible for its improved physical property¹⁴⁾.

CONCLUSIONS

Three non- γ_2 amalgams made from a dispersion-strengthened lathe-cut alloy (DA), the dispersion-strengthened spherical alloy (SD) and the Cu-In-added single composition spherical alloy (IL) and the conventional spherical alloy amalgam (SS) were compared by examining with a metallurgical microscope, electron probe microanalyzer and two types of microhardness tester. The findings were as follows:

1) The γ_2 phase in the matrix was remarkable in the SS amalgam but scarce in the DA, SD and IL amalgams, in which the matrix was markedly harder.

2) The residual alloy particles were composed of Ag and Sn in the SS, DA and SD amalgams but they were composed of Ag, Sn, Cu and In in the IL amalgam which was the hardest.

3) In the DA and SD amalgams, the hardness of the dispersant particles (Ag-Cu and Ag-Cu-Sn) was comparable to that of the residual alloy particles (Ag-Sn) and the Cu-Sn reaction phase generated around the dispersant particles was harder than any of the other phases.

4) In the IL amalgam, the Cu-Sn reac-

tion phase was found as minute granules scattered in the halo-like areas around the residual alloy particles.

5) Compared with the SS amalgam, the overall hardness was 20% higher in the DA and SD amalgams and 70% higher in the IL amalgam.

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LEGENDS FOR FIGURES

- Fig. 1. Scanning electron micrographs of the four types of amalgam alloy particles. SS: Conventional spherical alloy particles. DA: Lathe-cut alloy and spherical dispersant particles. SD: Spherical alloy and spherical dispersant particles. IL: Cu-In-added single composition spherical alloy particles
- Fig. 2. Optical microscopic structure of the four amalgams
- Fig. 3. Back-scattered electron images (top) and characteristic X-ray images of the constitutive elements of the four amalgams observed by electron probe microanalyzer