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# Characterization of Low and High Copper Amalgam Alloys and the Effect of Mixing Time on Their Physical Properties

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The available low and high copper amalgam alloys were characterized on the basis of their physical properties, as determined by A.D.A. Specification No. 1. The effect of mixing time on these properties were also examined. Sufficient mixing following the manufacturer's direction is required for lathe-cut alloys, regardless of the copper content.

Key Words: Low and high copper amalgams, Characterization in properties, Mixing time

## INTRODUCTION

At present, many amalgam alloys are commercially available. These alloys can be classified as having low and high copper contents and, also in terms of being lathe-cut, spherical or admixed. The various claims of the manufacturers concerning the advantages of their products in terms of both composition and particle shape, often causes confusion to the practitioner. Therefore, it is important to clarify the characteristics and performance of commercially available alloys. In this investigation, the physical properties of low and high copper alloys were examined mainly on the basis of the American Dental Association Specification No. 1[1,2].

## MATERIALS AND METHODS

1. Alloys and mixing procedure

Eighteen alloys were examined (Table 1). Mixing of alloy (0.6 g) and mercury was done mechanically. The manufacturer's recommended amalgamator, mercury to alloy ratio, and mixing time are shown in Table 2.

Specimens were tested according to the A.D.A. Specification No. 1[1]. Five specimens were made for each tests.

2. Examination of physical properties

1-hour and 7-day compressive strengths, 7-day diametral tensile strength, 7-day creep, 30-day Vickers hardness, dimensional change after 24 hours were examined.

Compressive and diametral tensile strengths were determined by a universal testing machine# with a cross-head speed of 0.2 mm/min.

Determination of creep was made following the A.D.A. Specification No. 1[1,2], using

# Type RS-2, Shimadzu Co., Japan

Table 1 Amalgam alloys applied

| Amalgam alloy | | Manufacturer | Type of products | Batch No. |
|---|---|---|---|---|
| Low copper | | | | |
| Lathe-cut | Luna 2 | G-C | powder | VR-11 |
| | Optaloy | L.D. Caulk | powder | 060677 |
| | Fluor Alloy* | Dentoria | capsulated | 1489 |
| spherical | Spherical 6 | Shofu | powder | 13 |
| | Spherical 8 | Shofu | powder | 527 |
| | Spherical 10 | Shofu | powder | 33 |
| | Hi Atomic M | G-C | powder | DO-31 |
| | Atomize Fine | Toyo | powder | 8012 |
| High copper | | | | |
| dispersant | | | | |
| (lathe-cut) | Dispersalloy, powder | Johnson & Johnson | powder | 1164-2 |
| | Dispersalloy, tablet | Johnson & Johnson | tablet | 19-JG |
| | Amalcap non gamma-2 | Vivadent | capsulated | 60616 |
| | Epoque 80 | Scania | powder | 7-090 |
| (spherical) | DP Alloy | Toyo | powder | 901-8013 |
| single compositional | | | | |
| (spherical) | Spherical D** | Shofu | powder | 017608 |
| | Dialloy | G-C | powder | JO-21 |
| | Lumi Alloy | G-C | powder | 070711 |
| | Tytin | S.S. White | capsulated | 387608 |
| | Sybraloy | Kerr | capsulated | 70912 |

* fluoride incorporated alloy ** Indilloy

a Micro Brinell hardness tester.#

A electrical micrometer## was used for testing of dimensional change during hardening, with a microscopic cover glass on the top of specimen. Measuring pressure was about 13 g.

Vickers hardness was obtained by means of a Micro Vickers hardness tester###, with a load and loading time of 50 g and 15 sec., respectively. Measuring procedure was described in detail elsewhere[3].

Each determination was made at 37°C, except for strengths. Creep and dimensional change was determined in an air conditioning oven and, determination of Vickers hardness was made on a specially designed heat plate[3]. Specimens for strength test were stored in an air conditioning oven, maintained at 37°C, until testing, although testing was performed under room temperature of 22±2°C.

\# Mori Shikenki Co., Japan

\## Type Minicom 30-J, Tokyo Seimitsu Co., Japan

\### Type M-2, Shimadzu Co., Japan

Table 2 Mixing procedure

| Amalgam alloy | Hg/alloy ratio | Amalgam mixer | Manufacturer's recommended mixing time (sec.) | Pestle or ball |
|---|---|---|---|---|
| Luna 2 | 1.0 | Hi-Mix | 15 | + |
| Optaloy | 1.0 | Silamat | 11 | – |
| Fluor Alloy | 1.2 | Shofu Amalgam Mixer D | 10 | – |
| Spherical 6 | 0.82 | Shofu Amalgam Mixer D | 10 | – |
| Spherical 8 | 0.82 | Shofu Amalgam Mixer D | 10 | – |
| Spherical 10 | 0.82 | Shofu Amalgam Mixer D | 10 | – |
| Hi Atomic M | 0.75 | Hi-Mix | 8 | – |
| Atomize Fine | 0.85 | Shofu Amalgam Mixer D | 10 | – |
| Dispersalloy, powder | 1.0 | Shofu Amalgam Mixer D | 10 | + |
| Dispersalloy, tablet | 1.0 | Shofu Amalgam Mixer D | 10 | + |
| Amalcap non gamma-2 | 1.2 | Silamat | 5 | – |
| Epoque 80 | 1.2 | Silamat | 5 | – |
| DP Alloy | 0.85 | Shofu Amalgam Mixer D | 10 | – |
| Spherical D | 0.86 | Shofu Amalgam Mixer D | 10 | – |
| Dialloy | 0.82 | Hi-Mix | 10 | – |
| Lumi Alloy | 0.83 | Hi-Mix | 15 | – |
| Tytin | 0.77 | Silamat | 2 | – |
| Sybraloy | 0.9 | Silamat | 10 | – |

## RESULTS

1. Physical properties of amalgams made according to manufacturer's instruction

1) 1-hour compressive strength (Fig. 1)

Large strength was noted with spherical alloys in general, with those of single compositional high copper ones being apparently greater. At the admixed high copper alloys, such large strengths was scarecely observed, indicating that the effect of particle shape is greater than that of copper content. Seven alloys showed smaller strengths than the requirement (80 MPa=816 kg/cm$^2$) of A.D.A. Specification No. 1[1]. The difference between minimum and maximum strengths was approximately 10 fold.

2) 7-day compressive strength (Fig. 2)

Although apparently large strengths were observed with high copper alloys, unlike those seen 1-hour compressive strength, the effect of particle shape was not so evident, i.e., single compositional high copper alloys exhibited a smaller strength than admixed high copper alloys. Low copper lathe-cut alloys had considerably lower strengths.

3) 7-day diametral tensile strength (Fig. 3)

The difference in strength between alloys was not as apparent as those of 7-day compressive strengths. Low copper alloys generally had a greater strength than high copper alloys. The influence of particle shape was not apparent.

4) Ratio of 7-day diametral tensile to 7-day compressive strength (Fig. 4)

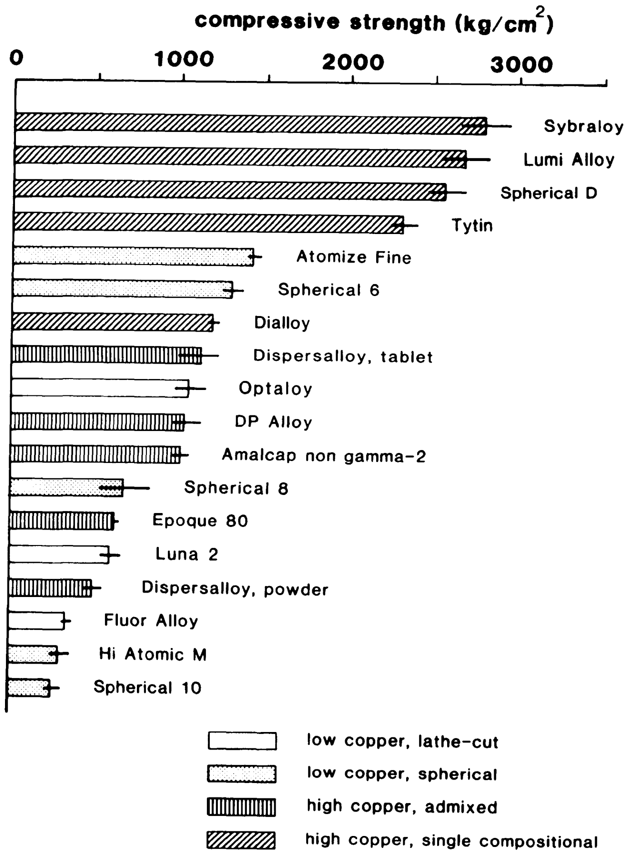


Figure 1 1-hour compressive strength

A higher ratio was seen in low copper alloys, findings which are converse to those for 7-day compressive strength. High copper alloys, had a lower ratio, were considered to be stronger but more brittle than low copper alloys.

5) 30-day Vickers hardness (Fig. 5)

Most of high copper alloys were considerably hard, especially single compositional ones. Little influence of alloy particle shape was observed at low copper alloys.

6) Creep (Fig. 6)

Most of low copper alloys exhibited considerably larger creep than high copper alloys. The influence of alloy particle shape was distinct, i.e., spherical alloys exhibited a small creep in comparison with lathe-cut alloys. Creep of applied alloys in this study satisfied the requirement (3.0%) of A.D.A. Specification No. 1[2], except for one. Difference in creep between alloys was 30 fold.

7) Dimensional change after 24 hours (Fig. 7)

Only 4 alloys, all admixed high copper alloys demonstrated expansion. Low copper alloys, especially those of spherical alloys, showed considerable shrinkage that exceed the range of requirement ($0 \pm 0.20\%$) of A.D.A. Specification No. 1[1], with 2 products.

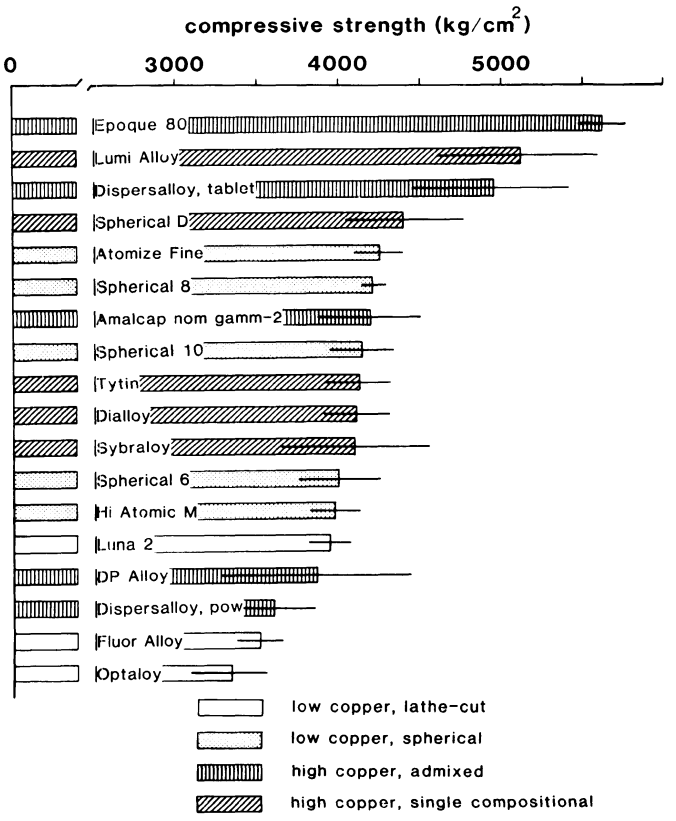


Figure 2 7-day compressive strength

2. Effect of mixing time on the physical properties of representative 15 alloys

1) 7-day compressive strength (Fig. 8–11)

A considerable decrease in strength was seen following the brief mixing, regardless of copper content. As for the effect of alloy particle shape, it was quite apparent that lathe-cut alloys had markedly decreased strength. Although most alloys exhibited maximum strength following their manufacturer's recommended time, little increase in strength was obtained with some alloys following slightly prolonged mixing.

2) 7-day diametral tensile strength (Fig. 12–15)

A markedly decrease in strength was observed at lathe-cut alloys following the brief mixing, regardless of the copper content. This is the same as compressive strength.

3) 30-day Vickers hardness (Fig. 16–19)

For the lathe-cut alloys, apparently small hardness was observed following the brief mixing, regardless to the copper content. On the other hand, with prolonged mixing, slight increase in hardness was observed with lathe-cut alloys. However, it was not observed with spherical alloys, except for only a few products.

3) Creep (Fig. 20–23)

Generally, the influence of the brief mixing on the creep was not apparent, dissimilar

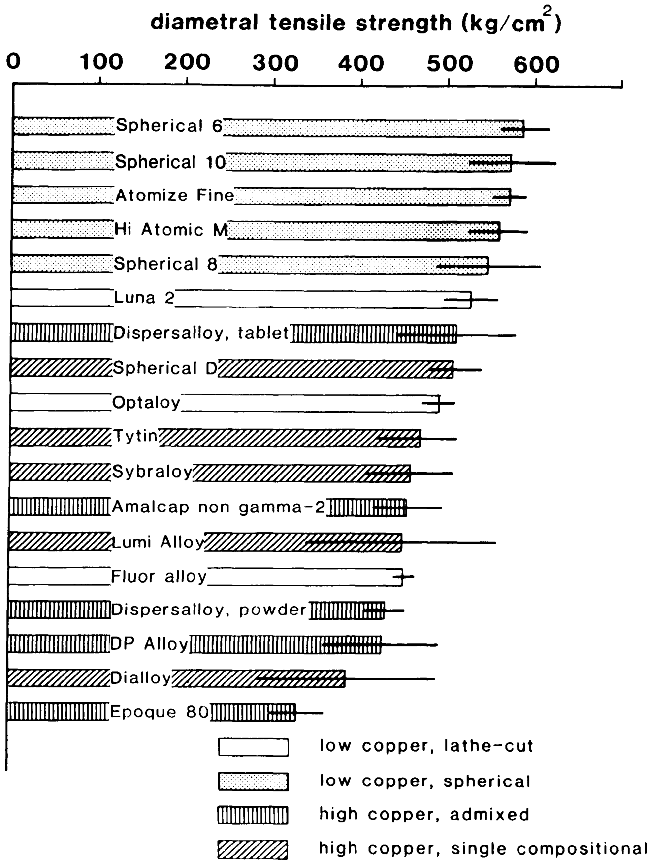


Fig. 3 7-day diametral tensile strength

to compressive and diametral tensile strengths. However, quite large creep was observed with lathe-cut alloys, regardless of the copper content. In the case of spherical alloys, although it was not apparent, slight increase in creep was observed following prolonged mixing. This was clear with low copper alloys, but not with high copper ones. In general, the high copper alloys, especially those of single compositional ones were less influenced by mixing time.

5) Dimensional change after 24 hours (Fig. 24–27)

Regardless of particle shape and copper content, brief mixing resulted in expansion or reduced contraction. This is especially apparent with lathe-cut alloys. On the other hand, with the prolonged mixing, all alloys exhibited an obvious contraction. This is more apparent at low copper alloys, regardless of particle shape. In general, spherical alloys, especially single compositional high copper ones, exhibited less influence of mixing time than lathe-cut alloys.

6) Comparison to manufacturer's recommended time (Fig. 28–31)

The effect of mixing time on the physical properties were expressed as a ratio to that of manufacturer's recommended time. 7-day compressive and diametral tensile strengths,

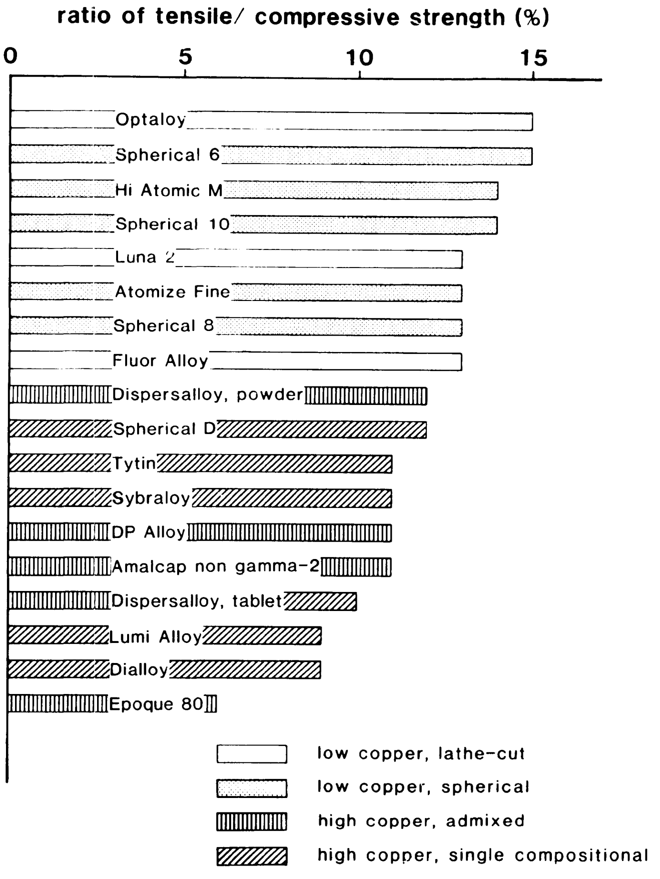


Figure 4 Ratio of 7-day diametral tensile to compressive strength

30-day Vickers hardness, creep and dimensional change was observed. Dimensional change was shown as a difference from that of manufacturer's recommended time. Lathe-cut alloys indicated more apparent influence of mixing time than spherical alloys, regardless of copper content. Among the physical properties examined, the most apparent influence was creep.

## DISCUSSION

Although many alloys are present on the market in this country, most of the recent products had a high copper content. Manufacturers have often claimed their rapid setting, large strength and hardness, small and stable dimensional change during hardening.

In regard to the large compressive strength and hardness at initial stage of hardening, it may be considered that such benefits exist, as less dangerous for marginal break with biting force even at the initial stage of hardening, chair time of patients would be reduced. However, some disadvantages also exist, such as difficult to restore the large cavity, inclusion of porosities at the marginal region of the restoration, because of a limited working

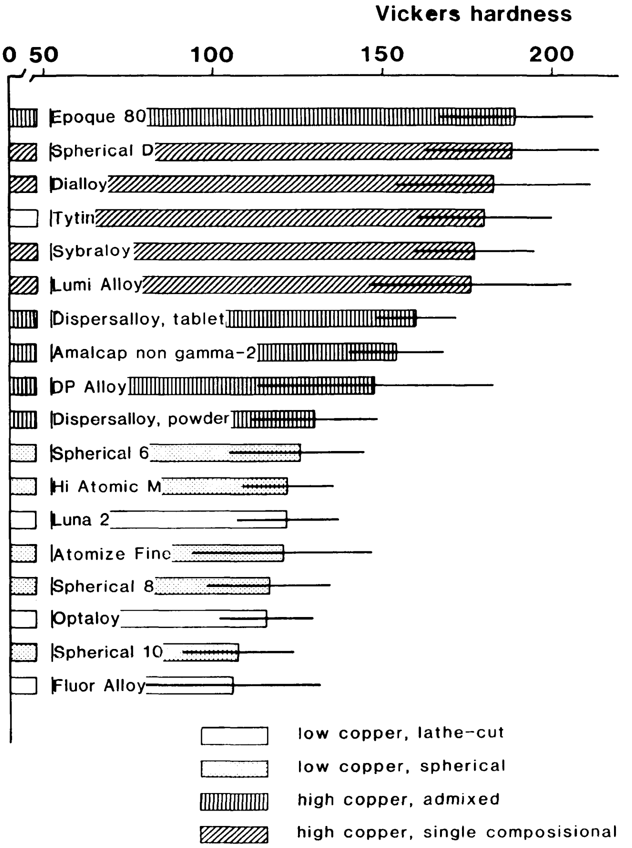


Figure 5 30-day Vickers hardness

time.

Although the compressive strength and hardness of recent alloys are larger than that of conventional ones[3,4], their clinical advantages have not yet been clarified. Some reports[5-7] have claimed that the large initial strength do not mean clinical benefits. Also, although it has been claimed that the smaller creep indicates little marginal fracture[8], recent investigation have clarified that there is no direct relationship between creep and clinical performance of restorations[9,10].

Dimensional changes during hardening are considered to be favourable and beneficial for clinical use, as reported elsewhere[11].

In respect to the effect of mixing time on the physical properties, apparently unfavourable results were obtained following brief mixing, especially with lathe-cut alloys. In contrast, a superior mechanical properties were obtained with slightly prolonged mixing. An apparent expansion was obtained with lathe-cut alloys following brief mixing. On the contrary, marked contraction was observed with low copper spherical alloys following prolonged mixing. The least influence of mixing time was observed with single compositional high copper alloys. In this study, following mixing with manufacturer's recommended time, slight contraction was generally obtained and only a few alloys showed

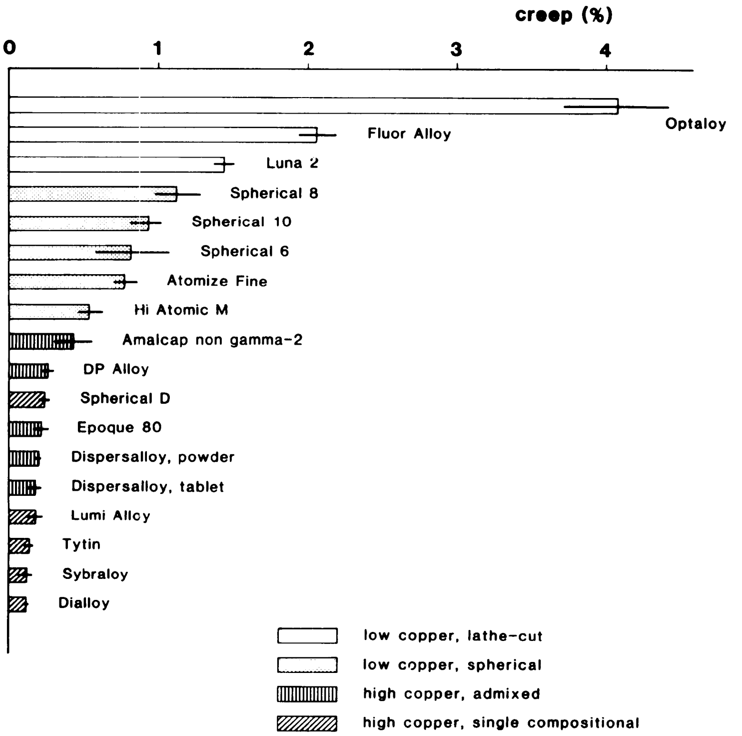


Figure 6 Creep

some expansion. Accordingly, it was confirmed that sufficiently mixed and condensed amalgams made from freshly marketted alloys would indicate slight contraction[12)].

The effect of mixing time on the microstructure of resultant amalgams were described elsewhere[13)]. The greatest influence was observed in regard to particle shape of alloys, supporting the above mentioned physical properties. Namely, it is quite apparent that the small strength, hardness, and large creep was caused not by the phases of amalgam itself, but by the enclosed porosities and spaces. Thus, it is considered to be important to reject the influences of such defects to estimate creep, because the creep is characteristic of the phase of amalgam itself[14,15)].

On the other hand, although a slightly superior mechanical properties may be obtained following prolonged mixing, its clinical benefits may be questionable. Severe heat generation, decrease in working time and shrinkage during setting are quite obvious. Probably, manufacturers of alloy determine the mixing time of their products from the standpoint of balancing the working time, mechanical properties and dimensional change. Thus, mixing of the amalgam should be made according to the manufactuere's direction, especially with the lathe-cut alloys.

The influence of mixing time on the properties of high copper amalgams is the same as on low copper ones. Since the mixing ability of high copper alloys are inferior than low copper ones[13,16,17)], careful mixing should be made, especially with admixed high copper alloys.

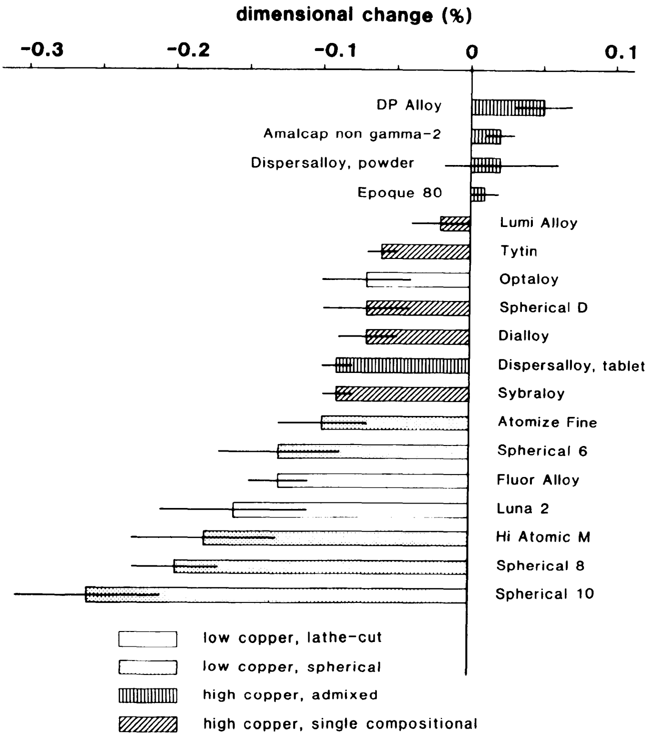


Figure 7 Dimensional change

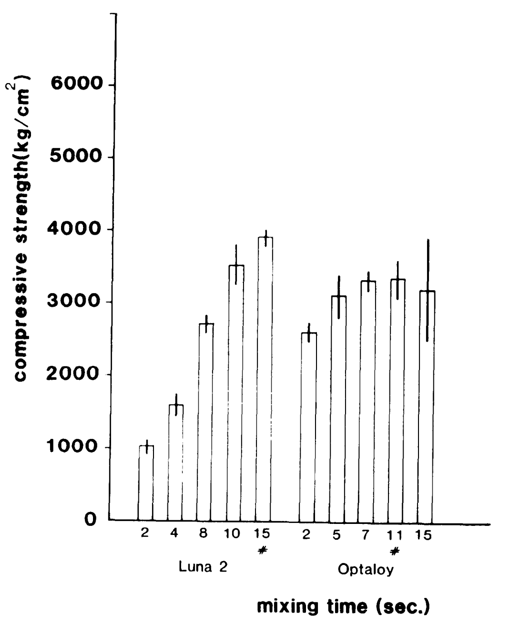


Figure 8 7-day compressive strength of low copper, lathe-cut amalgams

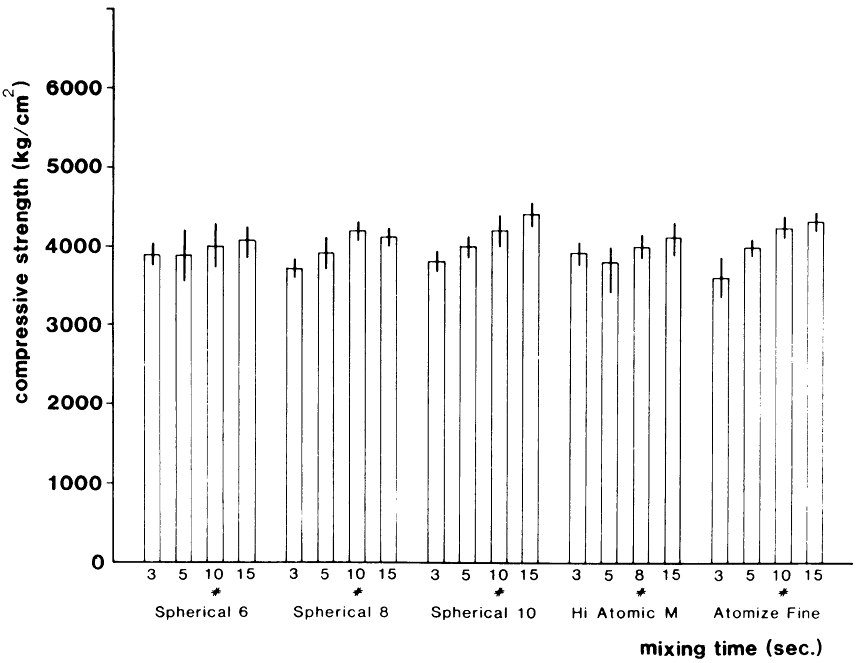


Figure 9 7-day compressive strength of low copper, spherical amalgams

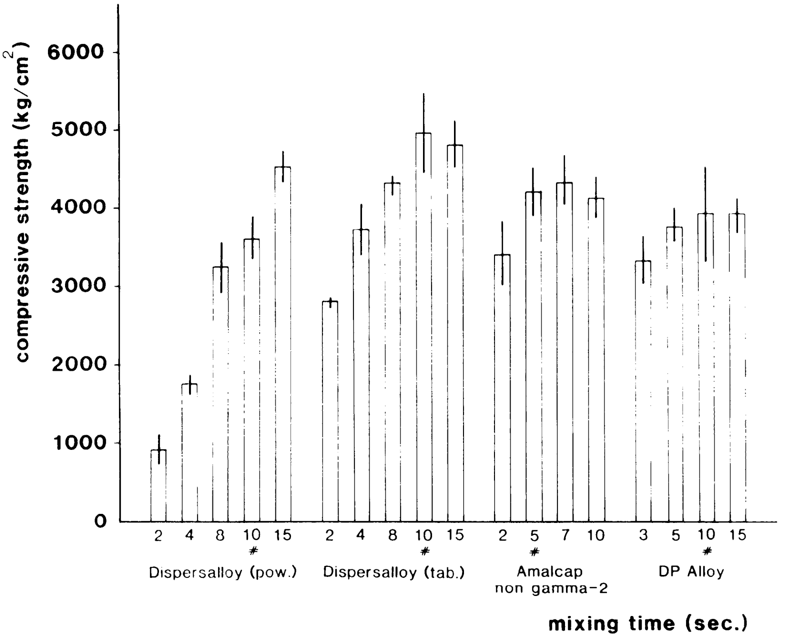


Figure 10 7-day compressive strength of high copper, admixed amalgams

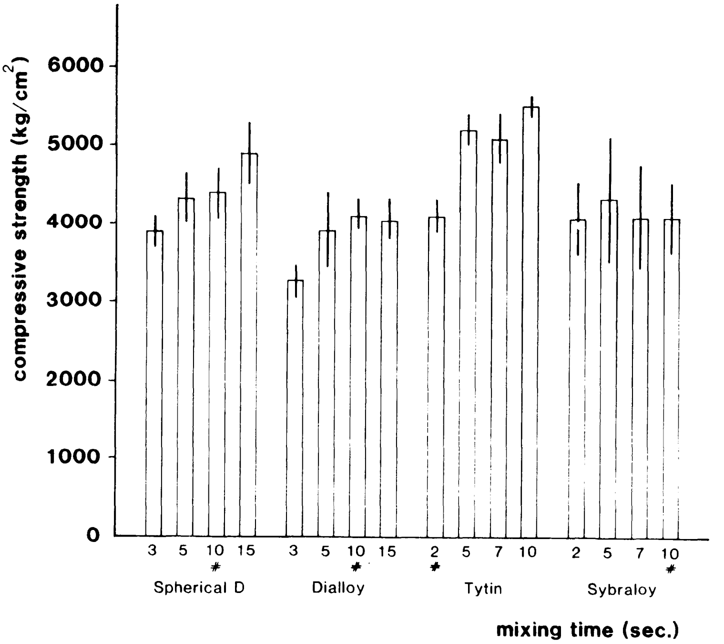


Figure 11 7-day compressive strength of high copper, single compositional amalgams

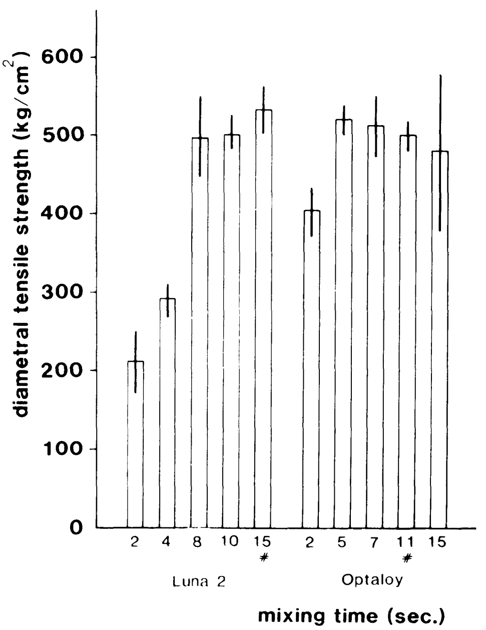


Figure 12 7-day diametral tensile strength of low copper, lathe-cut amalgams

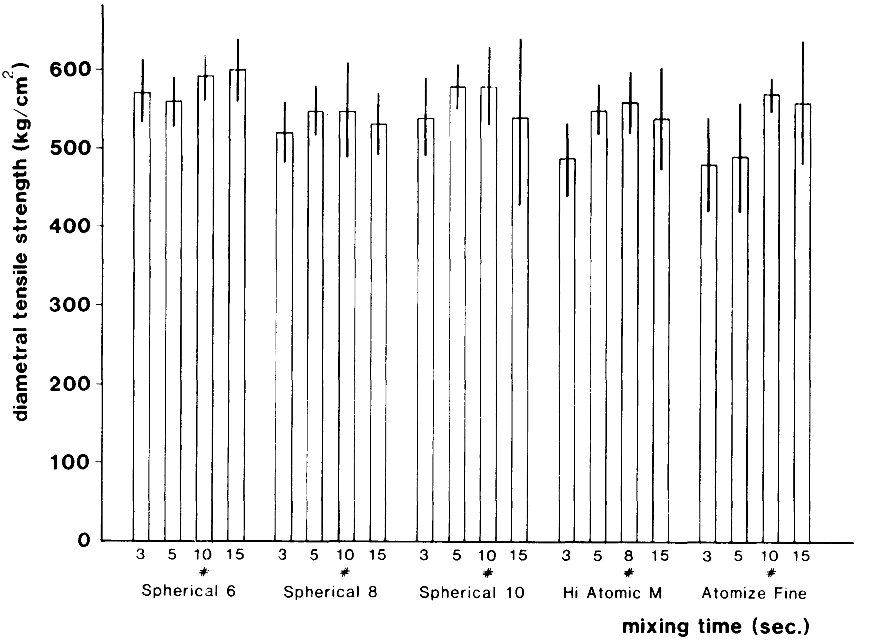


Figure 13 7-day diametral tensile strength of low copper, spherical amalgams

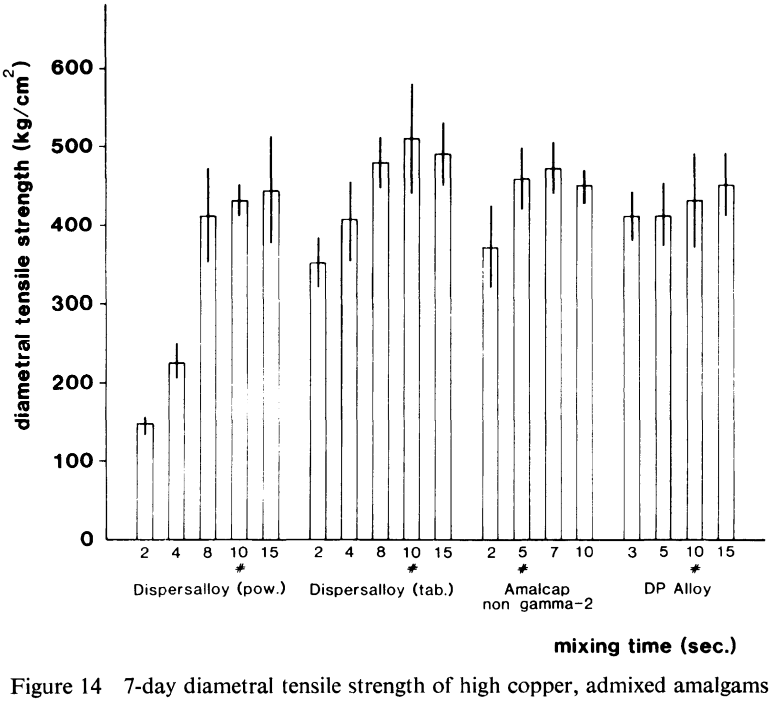


Figure 14 7-day diametral tensile strength of high copper, admixed amalgams

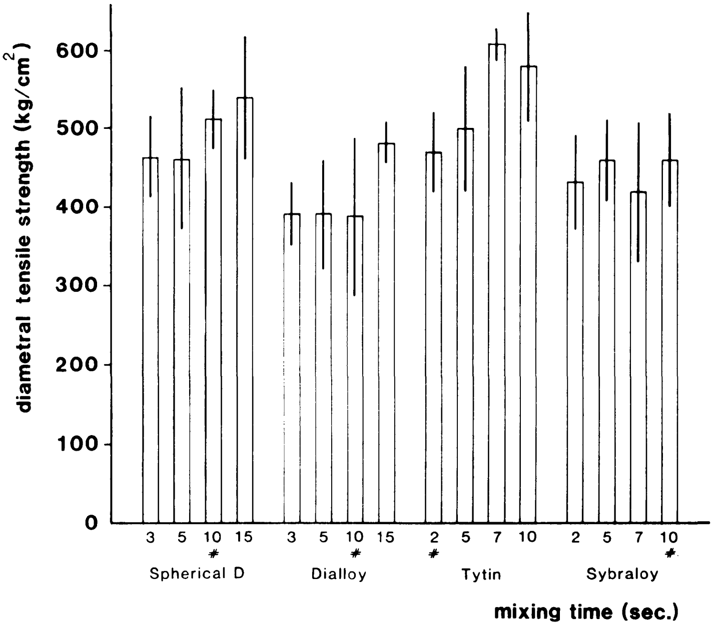


Figure 15 7-day diametral tensile strength of high copper, single compositional amalgams

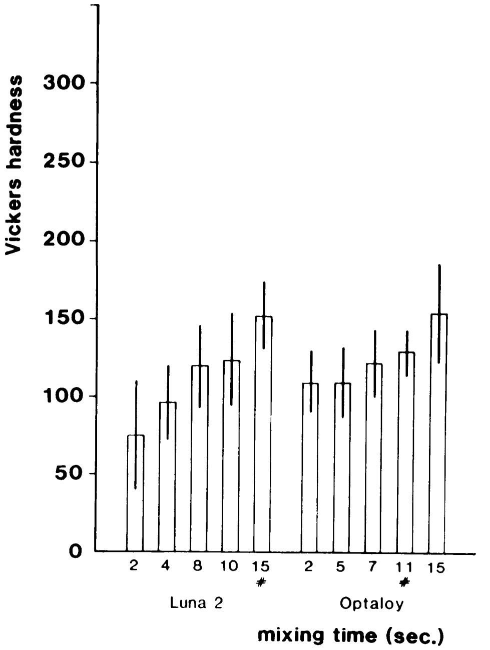


Figure 16 30-day Vickers hardness of low copper, lathe-cut amalgams

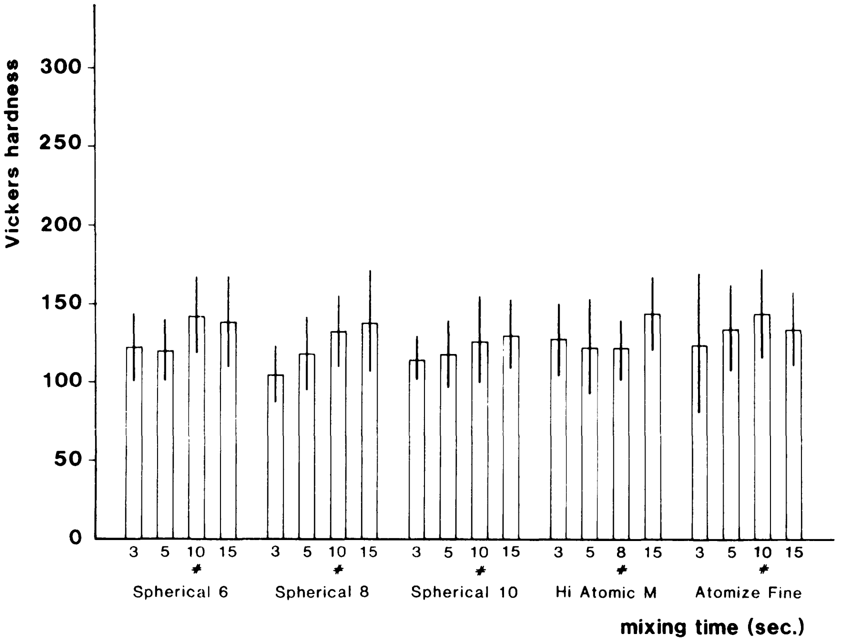


Figure 17 30-day Vickers hardness of low copper, spherical amalgams

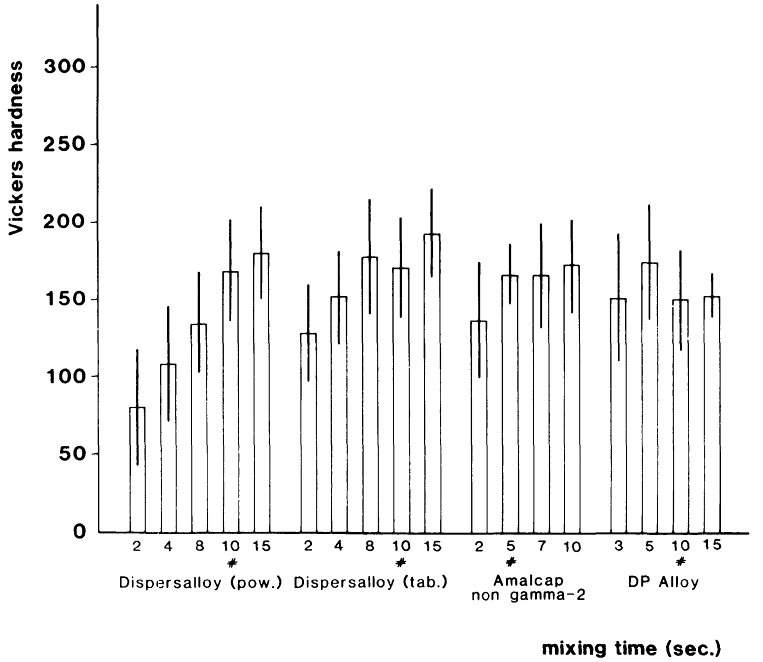


Figure 18 30-day Vickers hardness of high copper, admixed amalgams

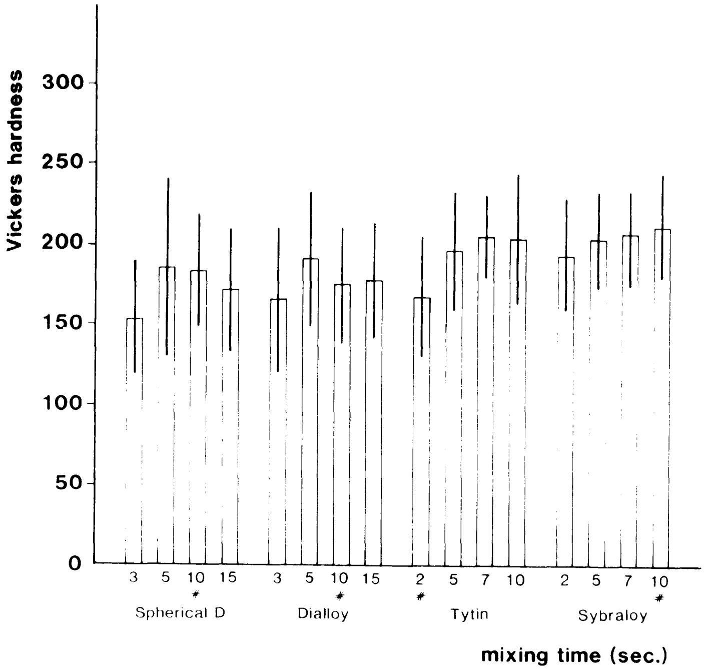


Figure 19 30-day Vickers hardness of high copper, single compositional amalgams

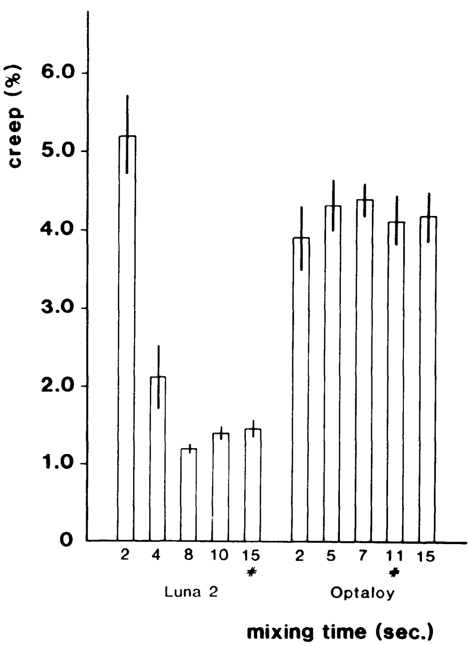


Figure 20 Creep of low copper, lathe-cut amalgams

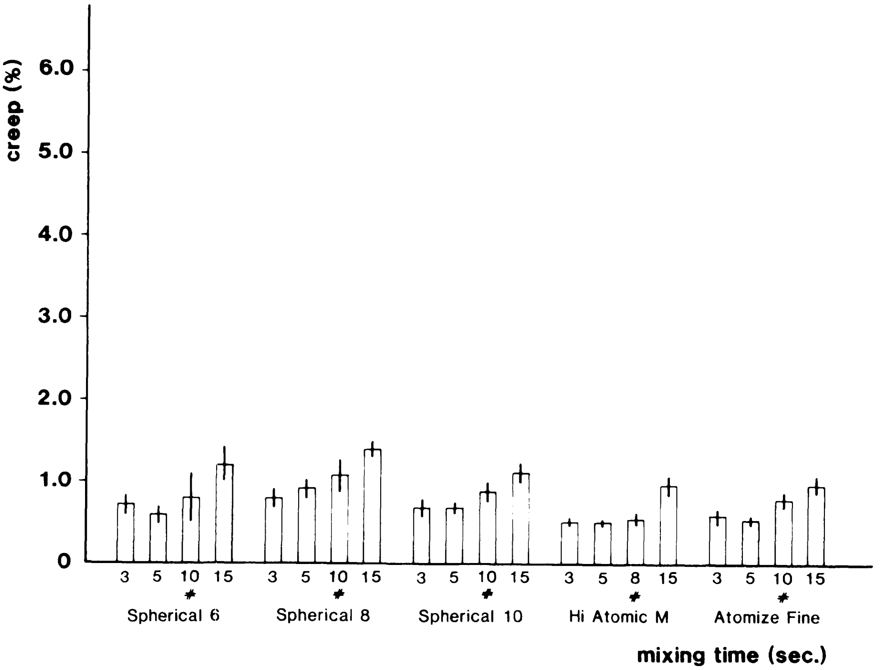


Figure 21 Creep of low copper, spherical amalgams

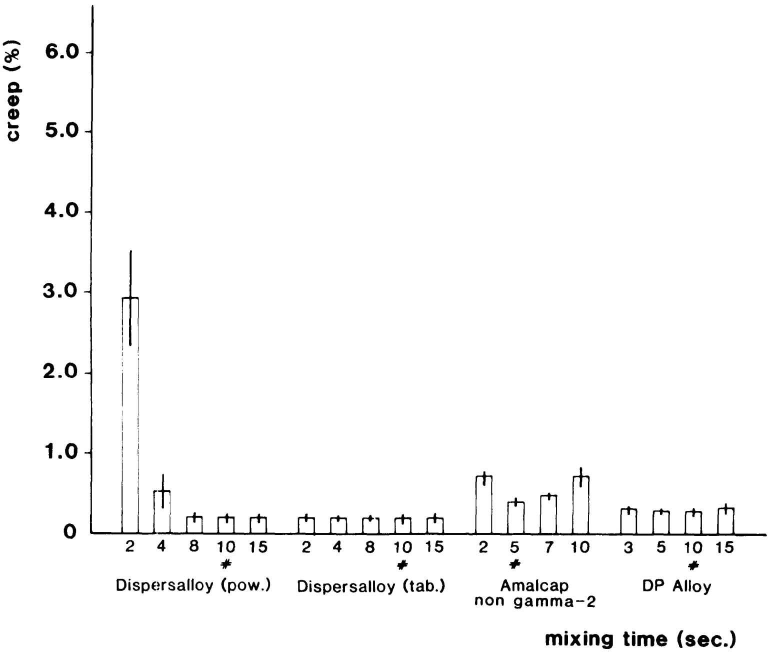


Figure 22 Creep of high copper, admixed amalgams

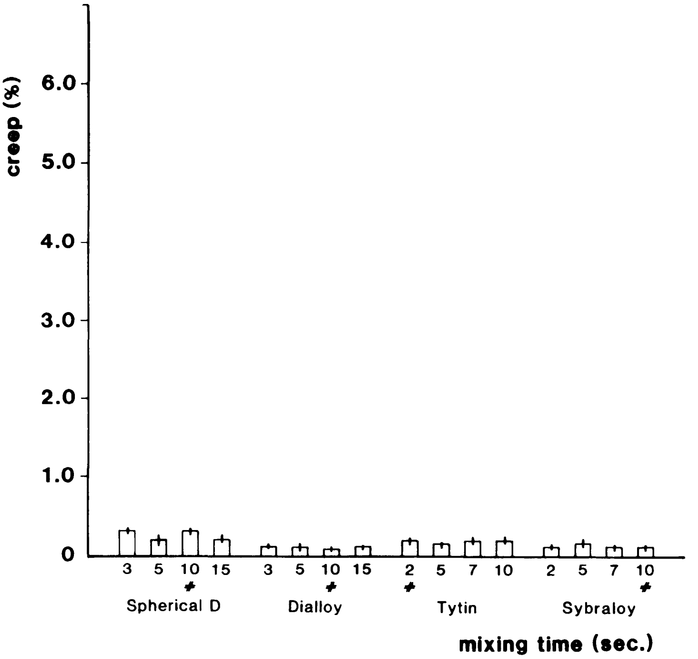


Figure 23 Creep of high copper, single compositional amalgams

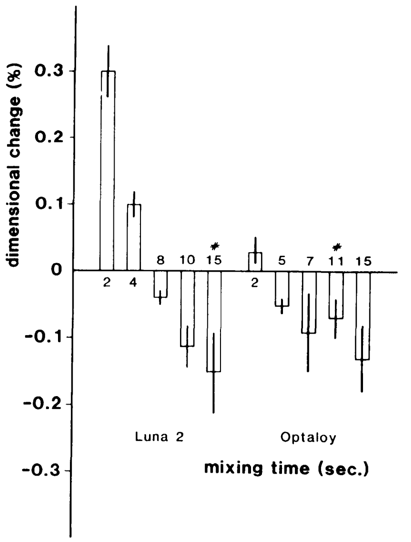


Figure 24 Dimensional change of low copper, lathe-cut amalgams

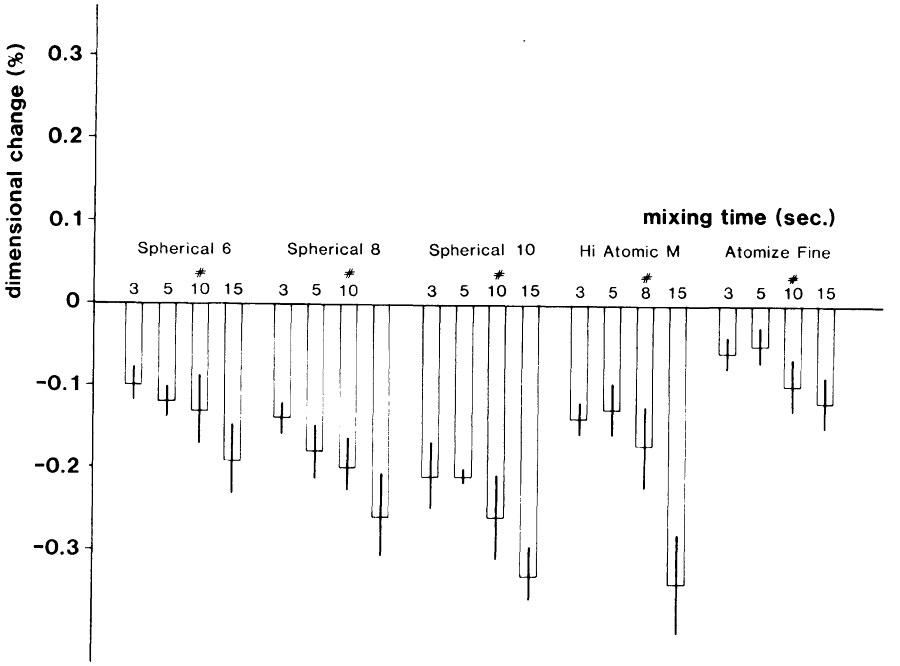


Figure 25 Dimensional change of low copper, spherical amalgams

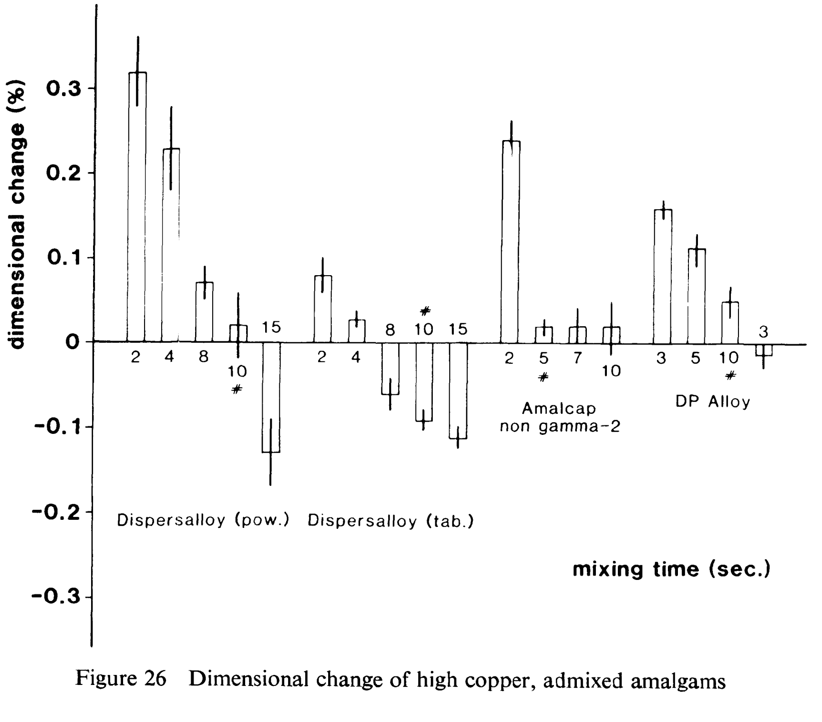


Figure 26 Dimensional change of high copper, admixed amalgams

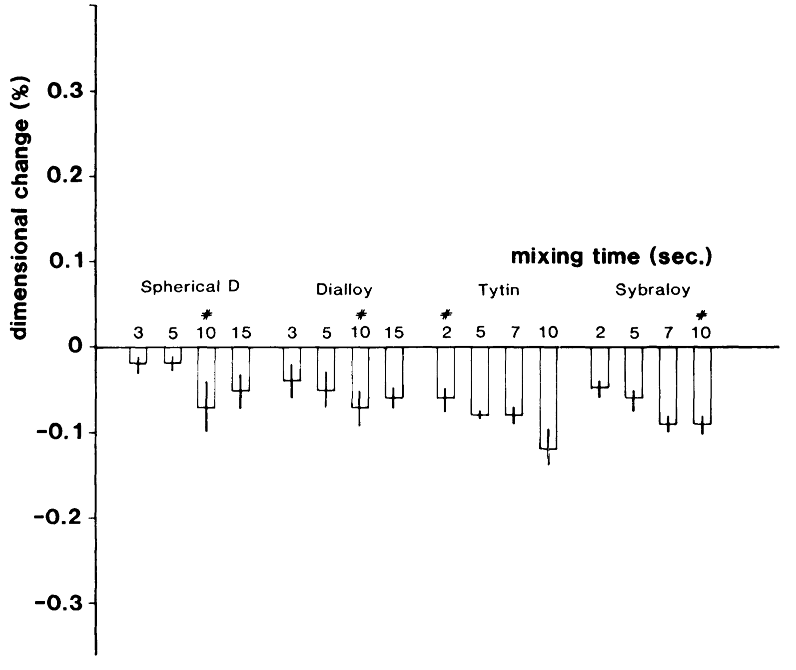


Figure 27 Dimensional change of high copper, single compositional amalgams

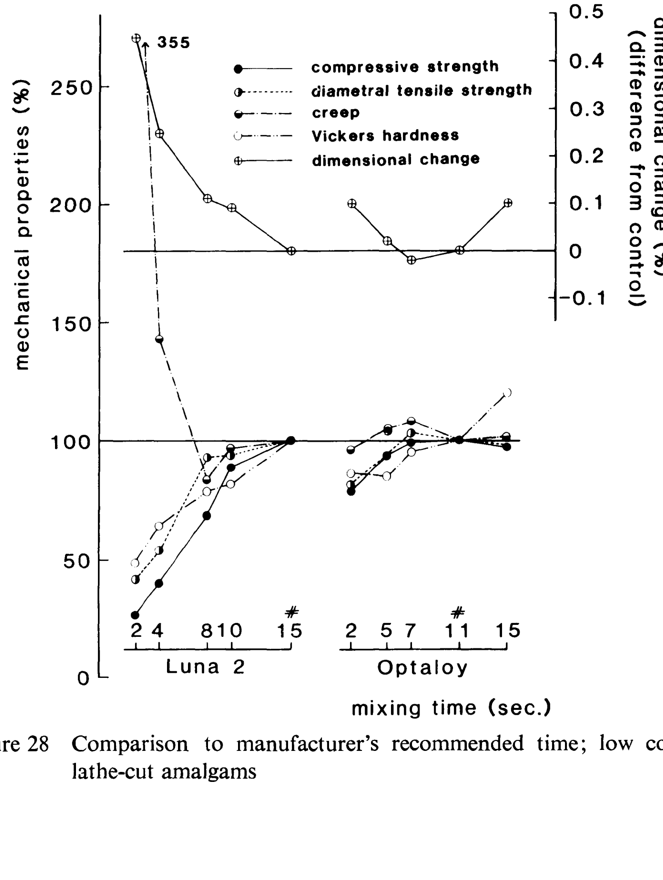


Figure 28 Comparison to manufacturer's recommended time; low copper, lathe-cut amalgams

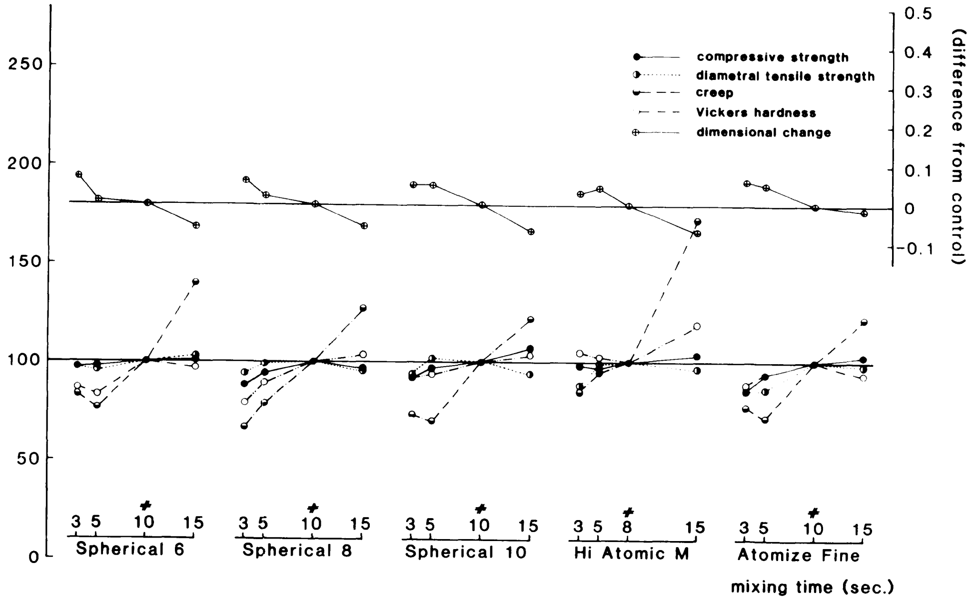


Figure 29 Comparison to manufacturer's recommended time; low copper, spherical amalgams

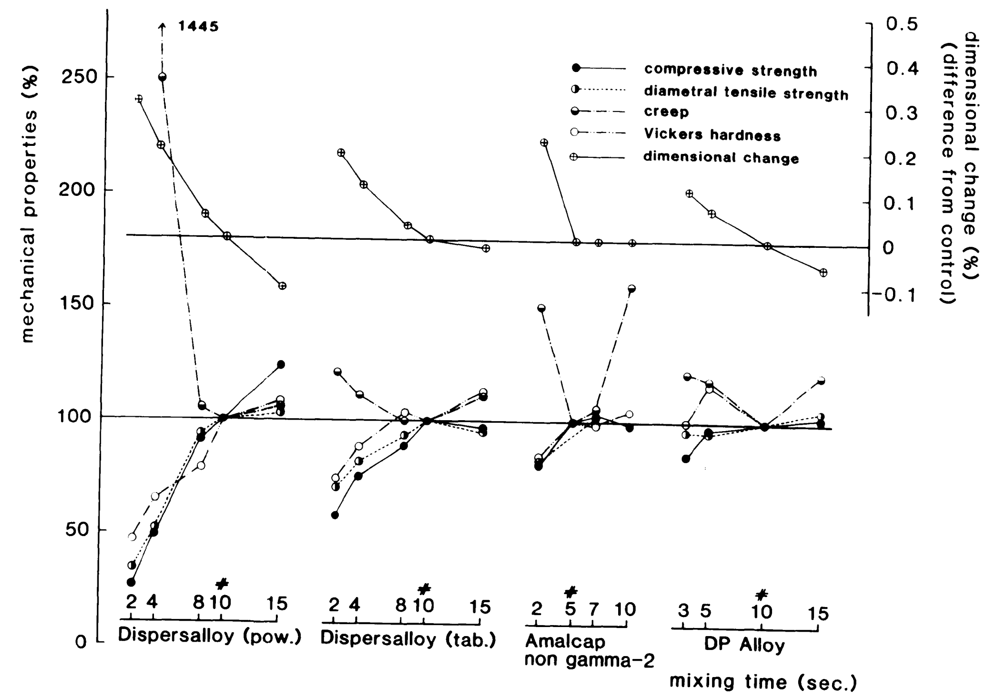


Figure 30 Comparison to manufacturer's recommended time; high copper, admixed amalgams

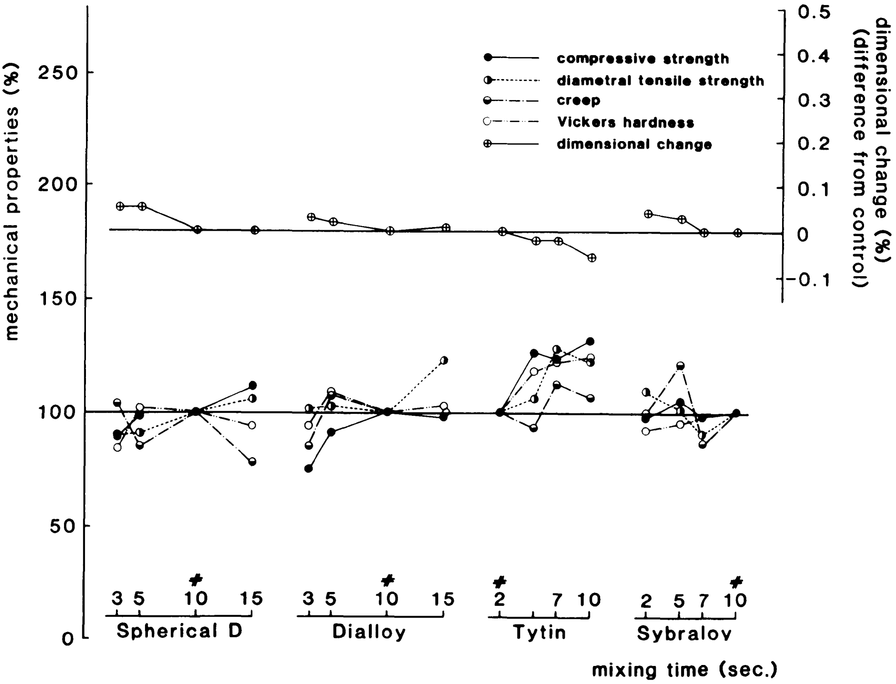


Figure 31 Comparison to manufacturer's recommended time; high copper single compositional amalgams

## CONCLUSIONS

An attempt was made to characterize the amalgam alloys presently on the market, low and high in copper content. Effect of mixing time on the physical properties were also investigated. Following results were obtained:

1. Although a great majority of the alloys recently appeared on the market of this country is of high copper content, distinctive differences in physical properties exist between single compositional and admixed ones. Namely, 1-hour compressive strengths of single compositional high copper alloys were extremely larger than those of low copper and admixed high copper ones. Yet, at 7-day compressive strength, these differences were not apparent.

2. In regard to 7-day diametral tensile strength, low copper, especially spherical particle, alloys had large strength. Ratio of 7-day diametral to 7-day compressive strength of low copper alloys was larger than that of high copper, especially single compositional, alloys.

3. Large 30-day Vickers hardness was observed at high copper, especially single compositional, alloys. Low copper alloys had a small hardness, regardless of its particle shape. Thus, high copper alloys are strong, hard, but brittle in nature, compared to low copper ones.

4. Regardless of the particle shape, low copper alloys exhibit a large creep. In contrast, an obviously small creep was obtained at high copper, especially single compositional,

ones.

5. Most alloys showed some contraction upon setting, after 24 hours. Admixed high copper alloys showed slight expansion and single compositional high copper ones had the least dimensional change.

6. Following the brief mixing, obviously inferior physical properties were observed with lathe-cut alloys, regardless of copper content. In contrast, such an unfavourable effects were scarecely observed with spherical ones, especially single compositional high copper.

7. With prolonged mixing, although unfavourable effect was scarecely observed at physical properties unlike to brief mixing, such shortcomings as heat generation, decreased working time, shrinkage were apparent. Thus, it is considered that only a little clinical benefits would be obtained by prolonged mixing.

8. Therefore, sufficient mixing following the manufactuere's direction is needed upon the practical use of lathe-cut alloy, regardless to the copper content.

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値が赤外炉焼成物において高値を示し，他の測定値はすべて赤外炉焼成物が低値を示した。また COLOR TABLE と各陶材焼成物とは色彩において両者の相違が見られ，白金炉と赤外炉などの熱源によっても陶材焼成物に色調の変化が表われた。

## サイクリック・クリープのコンピューターシミュレーション

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臨床で認められているように，義歯床の破折の大きな原因の一つは疲労である。これを究明するために，著者らはサイクリック・クリープ・テスタを設計製作し，その試用結果を先に報告した。

さらに本研究では，サイクリック・クリープの数値解析として，FEM をコアとするシミュレータを開発し，パーソナルコンピュータを使用してシミュレーションを行った。

試験方法として，JIS 及び ADAS に基づく片振り曲げ負荷を採用した。材料はエンジニアリングプラスチックの内から PEEK を採りあげた。この材料の DENTURE 使用時を想定してシミュレーションを行い，二，三の興味ある結果を得たので報告する。

なお，本研究は歯科領域における CAE としての試みであるが，なお機器の面で増強の予定がある。

## 各種歯科用アマルガムアロイの特性と，練和時間がその物理的諸性質に及ぼす影響

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入手可能な低，高銅型アマルガムアロイの物理的諸性質を主として A. D. A. 規格 No. 1 の方法によって測定してその特性を比較，考察した。同時に，練和時間の変化がこれらの物理的諸性質に及ぼす影響についても検討した。

低銅型アロイにおいては粒状が物理的諸性質に及ぼす影響は比較的小さかったが，高銅型アロイでは著明であった。即ち，球状粒子のみで構成されている単一組成型高銅アロイは他のすべてのアロイに比べて際立った特徴を示した。即ち，低銅型アロイは勿論，配合型高銅アロイも明らかに異なった特徴を持つことがわかった。

練和時間が物理的諸性質に及ぼす影響についてみると，不充分な練和は低，高銅型を問わず削片状アロイの性質に著しい悪影響を及ぼすが，球状アロイにはその影響が比較的小さいことがわかった。製造者の指示よりも過剰に練和した場合は機械的性質は若干向上するが，著しい発熱，操作時間の短縮，硬化時の収縮を伴なうので臨床的に利点があるとは思われなかった。物理的諸性質のうちで練和時間の影響を特に著明に表わしたのは，クリープであった。

## CuPd-AuCu 擬 2 元合金の規則化過程と時効硬化機構

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CuPd-AuCu 擬 2 元合金の規則化と時効硬化機構を検討した。e/a 値（伝導電子濃度）が0. 87より小さい合金では時効初期に短範囲規則化が進行し，その後不均一規則化機構により長範囲規則化が結晶粒界から起こった。これらの合金の時効硬化は CuPd 相または AuCu I 相の体積率の増加によるものであった。一方，e/a 値が