Effect of Copper Content Addition to Dental Amalgam Properties

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Abstract

A set of high-copper amalgam alloys have been prepared based on the change in the ratio of copper to silver using elemental components of high purity 99.9%. The amalgamation processes were done by mixing alloy powders with mercury at a fixed ratio. Structural properties were studied using X-ray diffraction and Optical Microscopy. Also, micro-hardness, and compressive strength were used to study some other important mechanical properties. The prepared amalgams were compared with well-known commercial amalgams; ANA 2000 and Standalloy F. The results of X-ray diffraction showed several prime phases in alloys and amalgams whose proportions and distribution depended on the copper content in the alloy. The results of mechanical test measurements showed a linear increase in the mechanical properties with increasing copper content in the amalgams. The results were similar to the measured values of the commercial amalgam.

Key Words: Dental Amalgam, X-ray Diffraction of Amalgam, Amalgam Hardness, Amalgam Compressive Strength.

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Introduction

Dental amalgam is a metallic alloy formed by reaction between an alloy powder and mercury. The alloy powder form containing silver, tin and copper along with other metallic elements added to improve physical and mechanical properties. Alloys powder can be classified into two groups: low-Cu content or conventional composition (5% or less copper) and high-Cu content (6 to 30% copper) [1-2].

Despite the early discovery of dental amalgam, it is still a competitive material for many newly discovered materials due to many reasons including its ease of use, low cost, long-term resistance properties and other features. The Silver-Tin-Copper alloy is one of the most important and widely used alloys in restorative materials as it has distinct physical and chemical specifications that makes it one of the leading materials in the field of dental material applications. Dental Amalgam consists of a combination of mercury with (Ag-Sn-Cu) alloy particles and has been used for several decades in the field of dental restoration. Due to the effect of the shape of alloy particles on its general specifications there are two types; lathe-cut particles and spherical particles. Many studies have been conducted on the shape of particles to improve their specifications and it was noted that some specifications have improved when the alloy powder is in the form of spherical particles [3-5]. Although liquid mercury-drawn powders have been used to produce dental fillings as reconstructive materials for dentistry for more than 150 years, dental amalgam filling is still the mainstay and widely used as reconstructive material for the treatment of back tooth decay, so there have been many attempts in recent years to improve the properties of dental amalgam [1, 6].

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Dental amalgam is still a favorite material where strength in amalgam is the key factor for selecting materials such as the restoration substance in dentistry [7].

The current work aims to improve some of the structural and mechanical properties of Ag-Sn alloy by adding different ratios of copper ranging from 14 to 26wt. % with a step of 4wt. %.

Materials and Experimental Procedures

Four sets of amalgam alloys were prepared that consisted of constant percentage of 30% Tin, while copper content is varying from 14 to 26% with interval of 4%. Copper is increased on the expense of silver content as shown in table (1) using conventional casting method. Also, two commercial alloys were used; the first with high copper content (ANA2000) and the other with low copper content (Standalloy F).

Table 1. The Nominal Components of the Alloys in Addition to Two Commercial Alloys.

Alloy	Ag [wt.%]	Sn [wt.%]	Cu [wt.%]
A	56	30	14
B	52	30	18
C	48	30	22
D	44	30	26
ANA	43.1	30.8	26.1
St-F	71	25.7	3.3

Amalgam alloys elements were mixed and melted at a temperature (1000-1100°C) using tube furnace type Carbolite. The operation was carried out under inert atmosphere using Argon gas. The alloy was left to cool down to room temperature. The alloy was reheated to a temperature (425-476 °C) for (17-19 hr.) to conduct the annealing process in order to obtain a homogeneous distribution of the γ-Ag_3Sn phase. The prepared cast alloys are subsequently transformed into powder using a grinder type Puluerisette 6. Next, the alloy powder was sieved to obtain a particle size between (40~70 μm). X-ray diffraction technique was used to show the phases present in different alloys using the Shimadzu XRD-6000 device. Dental amalgams were then prepared using high-purity mercury (99.9%) in accordance with weight ratios (1:1:2) using amalgamator device (Promedica) at vibrating rate (3600 vibr./min).

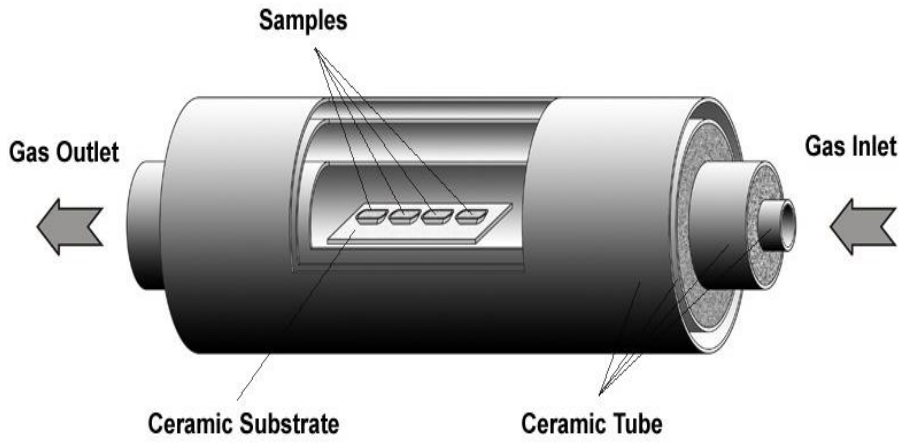

Figure 1. The Location of the Samples Inside the Tube Furnace.

In order to measure the compressive strength, the amalgam mass extracted from the amalgamation device was placed in acrylic mould and manually pressed with a pressure force equivalent to (2~3 MN/m^2), after one hour of hardening, it was removed from the moulds and now ready to be tested for compressive strength using Instron-1195 device. The samples were cold mounting using Polyester Resin and a hardener and then polished with Silicon Carbide paper of gradient (200-1200 µm). The samples were further polished with a polishing cloth and diamond powder pastes, at which point the samples were ready to be tested for

the micro hardness using micro-hardness tester HVS1000 by adopting the Vickers method and studied the micro-structure using the Reflected optical microscope.

Result and Discussion

a. X-ray Diffraction Analysis

Figure (2) shows diffraction patterns of alloy powder for samples (A, B, C, D) where all samples consist of a mixture of four main phases β-Ag_4Sn, η-Cu_6Sn_5, ε-Cu_3Sn and γ-Ag_3Sn, their ratios vary depending on the copper content in the alloy. It was noted that in sample A, which is the lowest copper content, it consists primarily of the β-Ag_4Sn phase which can be confirmed by the presence of the highest-intensity peak that is located at Bragg's angle $2\theta = 40^o$ and it is associated with the γ-Ag_3Sn phase indicating its presence in approximately the same ratios. Moreover, the ($2\theta = 42^o$) indicates the presence of the ε-Cu_3Sn phase in small proportions but higher than the concentration of the η-Cu_6Sn_5 phase.

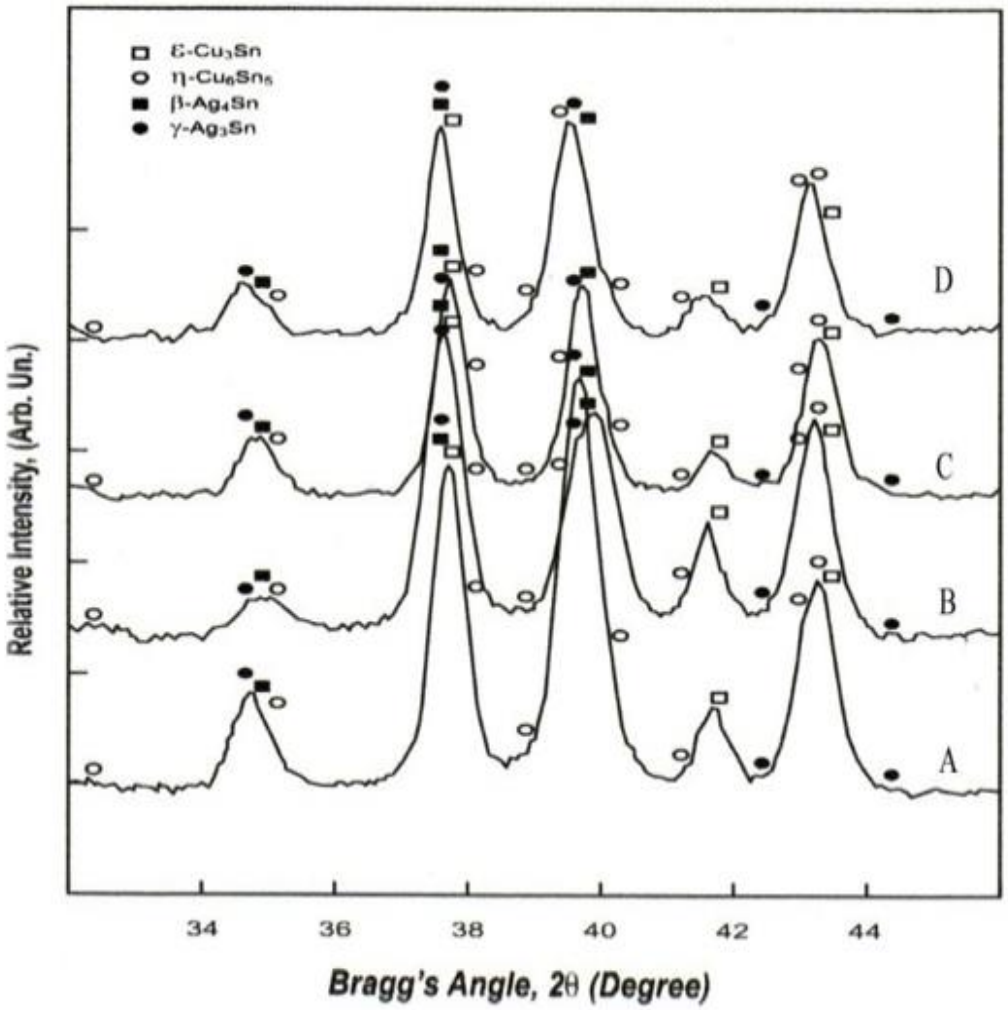

Figure 2. X-ray Diffraction Analysis of the Prepared Alloys.

The concentration of the four main phases change with the increase in copper content, as it is generally noted that the increased copper content increases the ε-Cu_3Sn and η-Cu_6Sn_5 phases at the expense of the β-Ag_4Sn and γ-Ag_3Sn phases (the latter phase is important and essential phase in the permanent tooth filler alloy as Silver increases strength while Tin reduces strength but increases the plasticity and setting time [3,8] which was confirmed by the decrease of the two peaks $2\theta = 38^o$ and $2\theta = 40^o$ that are jointly associated with the last two phases. These results are consistent with a number of studies in this field [9-10] which indicate that there is a significant tendency for copper to interact with Tin and thus cause a decrease in the amount of Tin available to interact with Silver to form the γ-Ag_3Sn and β-Ag_4Sn phases.

Clearly, the number of phases increased when the amalgamation process of alloys is conducted, as figure (3) shows at least six phases, namely the mercury alloys represented by the phase's γ_1-Ag_2Hg_3, γ_2-Sn_7Hg, and Ag_3Hg_2 as well as some non-interactive phases of the underlying alloy components. However, it should be noted from the figure that the γ_1-Ag_2Hg_3 has the highest percentage among the aforementioned phases, with the peak $2\theta = 38^o$ is considered to be the highest peak of the standard diffraction for this phase. It is also noted that the ε-Cu_3Sn phase is clearly increased with increased copper content in the alloy and this is confirmed by the peak $2\theta = 43^o$. The figure also shows that alloy A contains a small percentage of the phase γ_2-Sn_7Hg represented by peaks$2\theta = 44.5^o$ and $2\theta = 32^o$ in accordance with a number of literatures [3, 11], where this phase decreases or disappears when using high copper content in the all

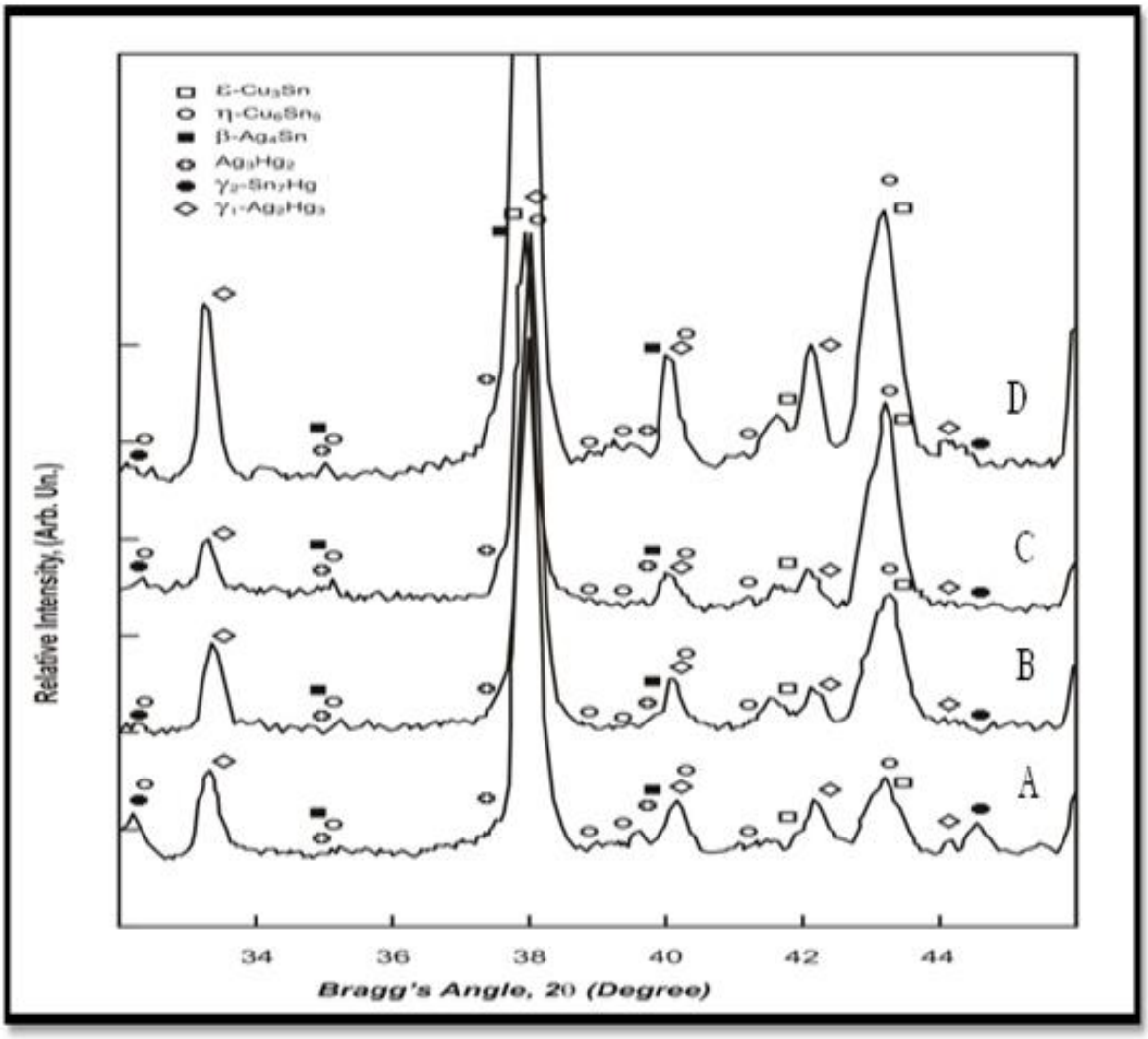

Figure 3. X-ray Diffraction Analysis of the Prepared Amalgams.

While figure (4) clearly shows the diffraction patterns for both commercial amalgams, it can also be observed the match between the commercial sample ANA and the prepared sample D, which have an almost equal copper content, whereas the sample St-F which has low copper content departs from the diffraction patterns of all prepared amalgams. The number of phases found in the commercial amalgam St-F is equal to that of the prepared amalgams. However, the distribution of phases differs significantly. For example, the phase γ_2-Sn_7Hg is present at relatively high ratios in amalgam; also, the ratios of the ε-Cu_3Sn and η-Cu_6Sn_5 phases are decreased while the Ag_3Hg_2 phase ratio increased.

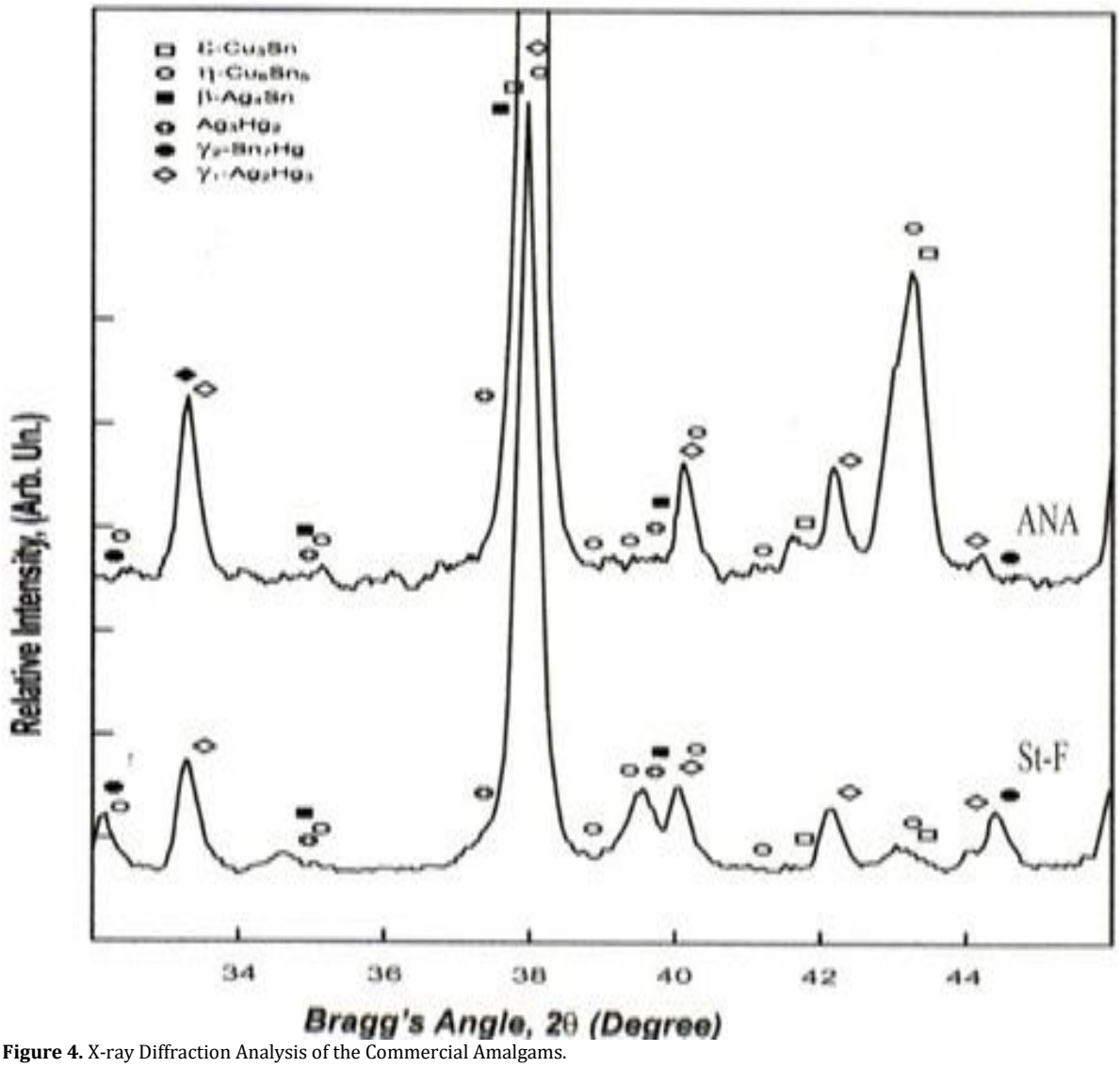

Figure 4. X-ray Diffraction Analysis of the Commercial Amalgams.

b. Microstructure Tests

The X-ray diffraction results for the prepared amalgams showed that they had a large number of phases of varying ratios and distributions depending on the copper content in the alloy and on the completion of the amalgamation process. These phases are formed by a mechanical mechanism called with triturating. During this process the Silver and Tin particles in the amalgam alloy spread towards the Mercury liquid that surrounds these particles, and it is expected that the crystals of the phase $_1\gamma$-Ag_2Hg_3 will be deposited around the particles of the amalgam alloy and then the process gradually continues until all the amount of Mercury used in the amalgamation process is consumed [12-13] also during this process other phases are deposited such as Ag_3Hg_2 or γ_2-Sn_7Hg while some phases remain non-reactive, such as ε-Cu_3Sn, η-Cu_6Sn_5 and β-Ag_4Sn. Thus, the microstructure for the results of the inverter optical microscope analyses can be determined by observing image (1). The amalgams shown in images A, B, C, D generally have four distinct regions, namely the light-colored areas representing the phase γ_1-Ag_2Hg_3, the gray granules representing the non-reactive β-Ag_4Sn phase that spreads randomly and dark areas, representing the ε-Cu_3Sn and η-Cu_6Sn_5 phases, as well as black spots which represent pores or gaps that occur as a result of trituration or the development of γ_1-Ag_2Hg_3 crystals.

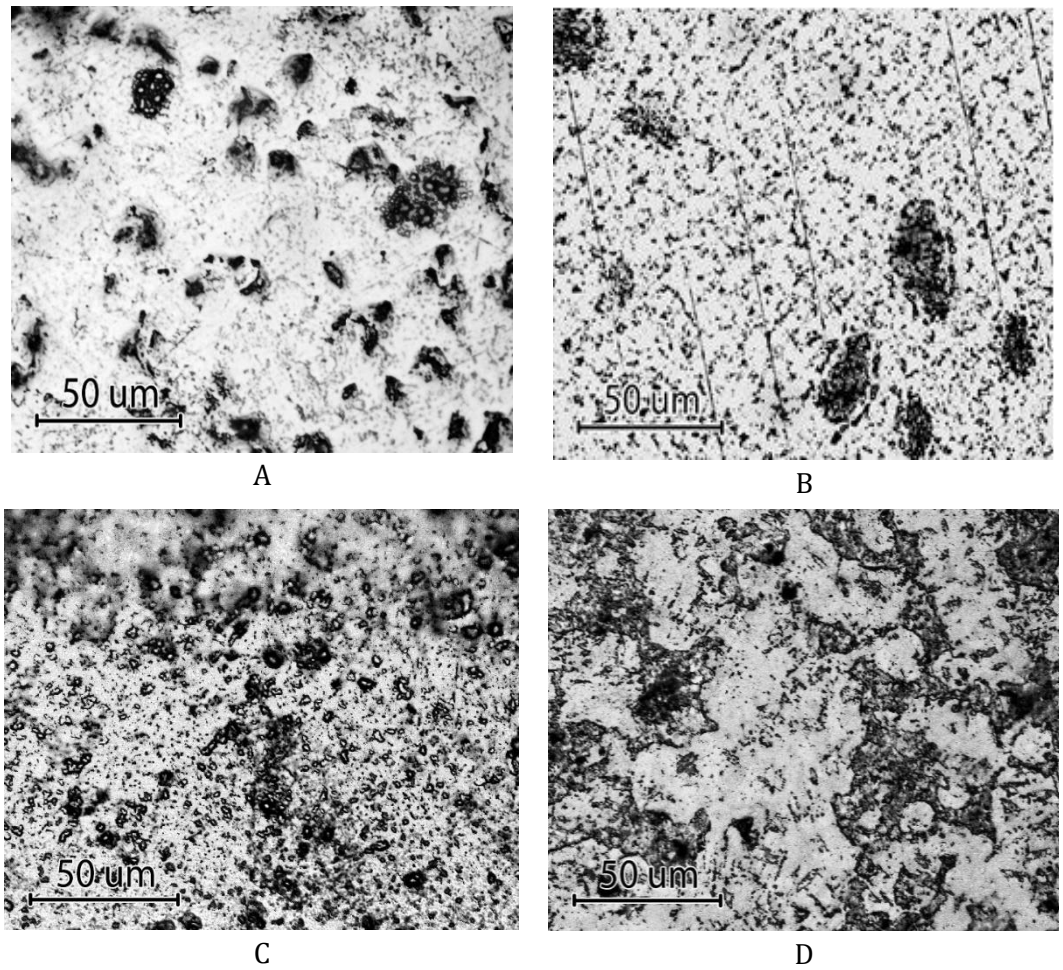

Image 1. Images of the Optical Microscope (Reflection Mode) for the Prepared Amalgamated Alloy

c. Micro Hardness Test

Figure (5) illustrates the hardness values of the prepared amalgams and commercial amalgams. The figure indicates a semi-linear relationship between copper content and the hardness values of the samples. Although hardness values increase slightly, the figure emphasizes that increase of copper content improves the mechanical properties of the amalgams and this is consistent with some studies in this field [5, 11, 14,15].

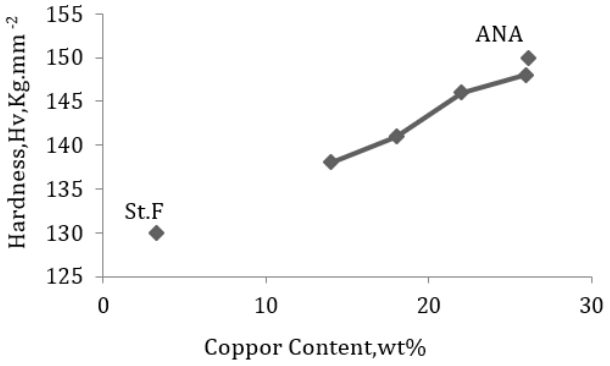

Figure 5. Hardness Variation with the Copper Content in the Amalgams.

It can be seen from Figure (5) that the commercial amalgam ANA has hardness slightly higher than the prepared samples, and this may be due to the difference in the microstructures between the two samples. On the other hand, the Commercial Sample St-F which has low copper content had reduced hardness, which confirms the importance of copper content to obtain amalgams with high mechanical properties.

d. Compressive Strength Test

Figure (6) shows a relationship similar to that obtained from the hardness measurement. Compressive strength values increase with the increase in copper content and this is consistent

with study in this field [15] and this can be explained by the fact that the increase in copper causes the removal of the phase γ_2-$Sn_{7\text{-}8}Hg$, which is weak phase in terms of mechanical properties [11]. The commercial amalgam ANA has slightly higher compressive strength than the prepared amalgams

while the sample St-F has low compressive strength.

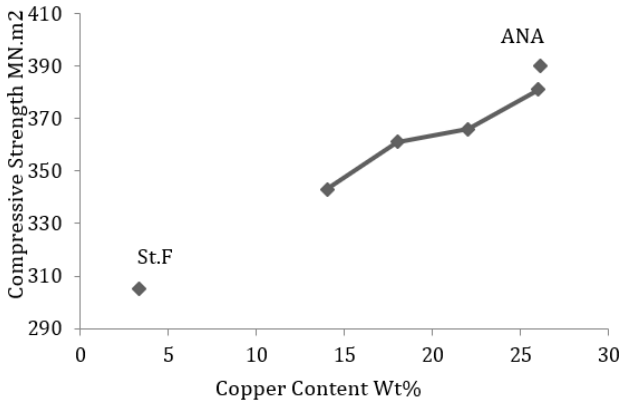

Figure 6. Compressive Strength Variation for Prepared and Commercial Amalgams with Copper Content.

Conclusions

A number of important conclusions can be drawn:

- The increase in copper in the Ag-Sn-Cu alloy, within the range used in this work, leads to a redistribution of the proportions of the resulted phases, but does not change the number of phases.
- The amalgam shows a linear increase in mechanical properties such as hardness and compressive strength with increased copper content in the alloy.
- Increasing copper content in alloy stabilizes the dimensional change of dental amalgam.

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