EFECTS OF COOLING RATE ON THE ELASTIC CONSTANS OF Cu$_3$Sn (ε) PHASE OF Ag-Sn-Cu DENTAL AMALGAM ALLOY
(Part II)

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Abstrak


Abstract

Effects of cooling rate (at the time of solidification) on the elastic constants of Cu$_3$Sn phase of Ag-Sn-Cu dental amalgam alloy were studied. In this study, three types of alloys were made, with the composition Cu-38.37 wt % Sn by means of casting, where each alloy was subjected to different cooling rate, such as cooling on the air (AC), air blown (AB), and quenched in water (WQ). X-Ray diffraction, metallography, and Scanning Electron Microscopy with Energy Dispersive Spectroscopy studies of three alloys indicated the existence of Cu$_3$Sn (ε) phase. Determination of the modulus of elasticity of Cu$_3$Sn (ε) phase was carried out by the measurement of longitudinal and transversal waves velocity using ultrasonic technique. The result shows that Cu$_3$Sn (ε) phase on AC gives higher modulus of elasticity values than those of Cu$_3$Sn (ε) phase on AB and WQ. The high modulus of elasticity value will produce a strong Ag-Sn-Cu dental amalgam alloy.
Introduction

Today, almost all of the amalgam for medical applications were made from high copper amalgam alloys (Ag-Sn-Cu alloys). In the high copper amalgam alloy most reactive element such as tin was completely utilized in the formation of Ag₃Sn (γ) and Cu₃Sn (ε) phases. Okabe et al had found that in the high copper amalgam alloy the Cu₃Sn (ε) phase particles were homogeneously distributed in a fine grain Ag₃Sn (γ) matrix. Therefore, it was predicted that the presence of Cu₃Sn (ε) phase could improve the mechanical properties of the amalgam alloy.

The mechanical property that is most often taken into consideration in the design and application of the high copper amalgam alloy is its elasticity. The modulus of elasticity of Cu₃Sn (ε) phase is regarded as an important and fundamental property, because a material with a low elastic modulus will readily elastically deform under functional stresses.

The modulus of elasticity of Cu₃Sn (ε) phase is also influenced by the manufacturing process, and a fairly high percentage of alloy is obtained through casting process. The performance of a cast product depends on its final cast structure, and the cast structure is dependent on the casting under which solidification of the casting taking place.

Ultrasonic wave applied to evaluate the elastic constant of materials was well established for a number of years. The ultrasonic method for determination of the elastic modulus of solid is base on the fact that the magnitudes of the velocity of longitudinal and transversal waves depend upon the elastic moduli of material.

Previous studies of high copper amalgam alloys have emphasized to study the microstructure and the mechanical properties of the Ag₃Sn (γ) phase. And at present very little emphasis has been placed on the microstructure and mechanical properties of Cu₃Sn (ε) phase. Therefore, the main objective of this study is to investigate the effects of the cooling rate on the modulus of elasticity of the Cu₃Sn (ε) phase.

Material and Methods

In order to obtain Cu₃Sn (ε) phase, three casting of 61.63 wt %Cu and 38.37 wt %Sn were melted in a graphite crucible using induction furnace. The melted alloy was poured into a mould with the dimension of 30 x 35 x 15 mm, and then the casting specimens were subjected to different cooling rate condition. The first casting specimen was allowed to solidify at room temperature (as cast specimen = AC), the second casting specimen was blown by air (AB), and the third was quenched in the water (WQ). The chilled layer that solidified in direct contact with the mould walls and the surface exposed on air was machined and cut off from the casting specimens. The final dimension of the test specimens were 25 x 30 x 10 mm.

The possible phases in the casting specimens were identified by X-ray diffraction technique, obtained with a Philips diffractometer (PW 3710) at 35 kV and 20 Ma operating condition. Specimens were scanned at 2°/minute from 20 to 100 degrees with Co kα radiation. The microstructures of these specimens were subsequently examined using standard metallographic technique on an optical microscope model PM 10D and Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM + EDS ; Philips 515).

The longitudinal wave velocity was determined by using an ultrasonic equipment (Universal ultrasonic flaw detector model USIP-12, Kraut-Kramer-Brandon) and a transducer (model U-Z-L Nortec) having a frequency of 5 MHz. The velocity of transmission of the transversal wave was measured with the same equipment and a couple transducer having a frequency of 4 MHz. The velocity of sound through the specimens were calculated using the following relationship.
\[ V_L = \frac{2T}{t} \]  

\[ V_T = \frac{T}{t} \]  

where \( V_L \) and \( V_T \) are the velocity of longitudinal and transversal waves (m/sec) respectively, \( T \) is the thickness of the specimens (mm) and \( t \) is the time between adjacent signals on the Oscilloscope (\( \mu \)sec).

The elastic constants of the specimen can be derived from the following equations:

Young's Modulus:
\[ E = \rho V_L^2 \left( \frac{3V_L^2 - 4V_L^4}{V_L^2 - V_T^2} \right) \]

Shear Modulus:
\[ G = \rho V_T^2 \]

Poisson ratio:
\[ \nu = \frac{V_L^2 - 2V_T^2}{2(V_L^2 - V_T^2)} \]

where \( \rho \) is the density of the specimen.

**Results**

The X-ray diffractograms of the air cooled (AC), air blown (AB), and water quenched (WQ) specimens are shown in Figure 1. The three specimens analyzed produce the same X-ray diffraction patterns result with indications of the orthorhombic structure of Cu-38.37 wt%Sn (\( \varepsilon \) phase), as evidence by the d values (Table 1).

The results of metallography analysis are shown in Figure 2. It can be seen that there are bright areas and also a darker areas as if it is consisting two phases. In addition, it can be observed that the faster the cooling rate, the smaller the microstructure produced. To insure whether the alloys produced consists of one or two phases, an analysis is carried out by using SEM and EDS. The results of SEM and EDS analysis (see Figure 3) shows that the dark area (Fig. 3, position 1) contains 39.17 wt% Sn, whereas the bright area (Fig. 3, position 2) contains 56.10 wt% Sn.

The calculation of the longitudinal and transversal waves velocity, the density, the modulus of elasticity, and the Poisson ratio of the AC, AB, and WQ Cu-38.37 wt%Sn can be seen in Table 2. The AC result of Cu-38.37 wt%Sn (\( \varepsilon \) phase produce a higher value of the modulus of elasticity than the modulus of elasticity of AB and WQ result of Cu-38.37 wt%Sn phases. These results indicated that the AC result of Cu-38.37 wt%Sn (\( \varepsilon \) phase is more homogeneous which is implied by the higher value of the density of AC result of Cu-38.37 wt%Sn phase compare to the density of AB and WQ result of Cu-38.37 wt%Sn phases.

**Table 1.** d spacing values (Angstrom), phase identification and the structure of As Cast (AC), Air Blown (AB) and Water Quenched (WQ) casting specimen of Cu-38.37 wt%Sn

![Table 1](image_url)

![Figure 1](image_url)

**Figure 1.** The X-ray diffraction patterns of Cu-38.37 wt%Sn specimens; (a) As cast, (b) Air Blown and (c) Water Quenched.
Table 2. The Density, The Longitudinal Wave Velocity, The Transversal Wave Velocity, The Young’s Modulus, The Shear Modulus and The Poisson Ratio of As Cast (AC), Air Blown (AB) and Water Quenched (WQ) casting specimen of Cu-38.37 wt%Sn.

<table>
<thead>
<tr>
<th>Cu - 38.37 wt% Sn</th>
<th>( \rho ) (kg/m³)</th>
<th>( V_L ) (m/sec)</th>
<th>( V_T ) (m/sec)</th>
<th>E (GPa)</th>
<th>G (GPa)</th>
<th>( \nu )</th>
</tr>
</thead>
<tbody>
<tr>
<td>As Cast</td>
<td>8932</td>
<td>4505</td>
<td>2383</td>
<td>132.5</td>
<td>50.7</td>
<td>0.31</td>
</tr>
<tr>
<td>Air Blown</td>
<td>8885</td>
<td>4506</td>
<td>2210</td>
<td>116.4</td>
<td>43.4</td>
<td>0.34</td>
</tr>
<tr>
<td>Water Quenched</td>
<td>8821</td>
<td>4402</td>
<td>2195</td>
<td>113.4</td>
<td>42.5</td>
<td>0.33</td>
</tr>
</tbody>
</table>

Figure 2. The micrographs of Cu-38.37 wt%Sn specimens; (a) As Cast, (b) Air Blown and (c) Water Quenched. Magnification 500 x.
Discussion

The range of Sn composition which is 36.10 - 39.17 wt% can still be classified within the Cu$_3$Sn (ε) phase formation area. In this way, the production of the alloys of Cu-38.37 wt% Sn AC, AB, and WQ produces a single phase of Cu$_3$Sn (ε) as has been shown by the result of X-ray diffraction analysis. It can be seen here, although the cooling rate is slowed down, the chemical equilibrium cannot be achieved. Consequently, the Cu$_3$Sn (ε) phase is produced with a little difference in the distribution of Sn.

According to Ambardar et al, in an alloy which is produced by means of casting cannot be free from defects, such as porosities, microcracks, and inclusion, although the solidification condition is maintained optimally in order to avoid the formation of porosities as a result of solidification shrinkage. In the alloy of Cu-38.37 wt% Sn which undergoes a fast cooling rate, especially the cooling rate by quenched in the water, there is a higher possibility of cracks in the structure due to rapid solidification rate, so that the modulus of elasticity is lower compare to AC alloy, although the microstructure is smaller than the AC alloy.

As has been stated before, the Cu$_3$Sn (ε) phase is the second constituent in the Ag-Sn-Cu dental amalgam alloy. As a second constituent, the Cu$_3$Sn (ε) phase is evenly and homogeneously distributed in the matrix of the Ag$_5$Sn (γ) phase which constitutes the main constituent in the Ag-Sn-Cu dental amalgam alloy. Based on the research conducted by Grenoble and Katz, it is found that the Young's modulus and the shear modulus values of the Ag$_5$Sn (γ) phase is 83.5 and 30.67 GPa respectively. The research of Grenoble and Katz was conducted by using ultrasonic technique and also with pressure, therefore, the values obtained constitutes the modulus of elasticity of the material with minimal porosity.

From this research it is found that the Young's modulus and the shear modulus values of Cu$_3$Sn (ε) phase produce either by AC, AB, or WQ are much higher if compare to the Young's modulus and the shear modulus of Ag$_5$Sn (γ) phase. Therefore, it can be stated that the Cu$_3$Sn (ε) phase plays an important role in reinforcing the structure of Ag-Sn-Cu dental amalgam alloy. Such structure of the Ag-Sn-Cu dental amalgam alloy can be analogous to the structure of particulate composite in which the Ag$_5$Sn (γ) phase is the matrix and the Cu$_3$Sn (ε) phase is the reinforcer.

Conclusion

The production of Cu-38.37 wt% Sn alloy with air cooling (AC), air blown (AB), and water quenched (WQ) yields a single phase of Cu$_3$Sn (ε) phase. The AC result of Cu$_3$Sn (ε) phase yields Young's modulus and shear modulus values higher than the AB and WQ result of Cu$_3$Sn (ε) phase, which implied that the microstructure produced from air cooling is more homogeneous. High values of the modulus of elasticity measured with ultrasonic technique indicated that the Cu$_3$Sn (ε) phase plays an important role in reinforcing the structure of Ag-Sn-Cu dental amalgam alloy.

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References