Improvement the Strength of a Dental Filling Restoration by Adding of NiTi alloy Powder to Galloy

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Abstract

Gallium has the second lowest melting point of all metals after mercury (29.8 ºC) and has the ability to react with metals and alloys at room temperature to produce a workable plastic mass that hardens with time. As a result Gallium-based alloys have been introduced to the dental market as mercury-free amalgam substitutes. Filling material produced by mixing amalgam alloy powder with gallium liquid instead of mercury. This work involves addition of NiTi alloy powder to Galloy to improve some mechanical properties. These properties included compression, hardness, tensile strength and creep. The characterization of prepared NiTi/Galloy with five wt% of addition (1, 2, 3, 4, and 5 wt%) was done by XRD and SEM/EDS. The results showed that the hardness, compression and diametral tensile strength were increased with increasing wt% of added NiTi alloy powder and the creep percent was decreased where NiTinano powder alloy disperses in the matrix of the alloy and prevents the dislocation movement, also the NiTi powder particles have unique properties of super elasticity that work to increase the creep strength.

Keywords: *Galloy; Amalgam; Dental alloy; NiTi alloy.*

Introduction

Amalgam is still a preferred material where strength is the requirement for material selection as restoration material in dentistry, but the toxic effect of Hg led to the development of gallium alloy [1]. As a result, introduction of Gallium based alloys have been occurred as mercury free amalgam which was suggested by Puttkamer as long ago as 1928 (2,3). On the other hand, Ni-Ti alloy was developed by W. F. Buehler in 1960s. A shape memory effect of this intermetallic alloy was found from its thermodynamic properties which controlled heat treatment [4]. This alloy was named Nitinol, an acronym for the elements from which the material was composed; ni for nickel, ti for titanium and nol from the Naval Ordnance Laboratory. This alloy has a lower modulus of elasticity and greater strength compared with stainless steel, therefore the advantage of using NiTi instruments during the preparation of curved root canals has been introduced, because the files will not be permanently deformed as easily as would happen with traditional alloys[5]. Many authors highlighted on Ga alloy for dental applications (6-11), but this alloy had some defects and many attempts have been done to improve this alloy.

The present work aims to improve some mechanical properties of Ga alloy by adding NiTi alloy powder with five percents (1, 2, 3, 4 and 5 wt%) to investigate the hardness, compression, tensile strength and creep.

Materials and Method

Materials: The elements and materials used in this work are gallium, indium, tin, and bismuth. Ga, In and Sn used for the preparation of the liquid alloy. A standard amalgam alloy used as powder alloy that mix with liquid alloy. All elements used in this work with high purity obtained from different origins.

Liquid alloy was prepared by alloying gallium, indium and tin together at 230ºC until all the elements be liquid and let it cool down to room temperature, the weight percentages of Ga, In and Sn were (64, 24, and 12 wt.%) respectively and the melting point of this alloy is -19ºC.

Preparation of Base Alloy: The preparation of specimens were done according to the ADA specification No.1[12] by trituration 1:0.5 g of powder alloy and liquid alloy respectively for preparation fills by amalgamator type (YDM-Pro), the trituration time is 8 seconds. Specimen dimensions were 6mm in diameter and 12mm in height as cylindrical shape. For specimen preparation, a Teflon mold was used and after the trituration, the fill paste was immediately put in the hole of the die by using a special instrument with the diameter 2.5mm called condenser, and then it was compressed into the die 14 MPa for 85 seconds.

The powder of NiTi with a weight percentage of (55% Ni + 45% Ti) has been wetly mixed by planetary automatic ball mill with five steel balls differ in diameter to mix and refine metal powder for three hours. Ethanol has been used as a mixing medium of wet mixing and then 5 g of NiTi powder mixture has been compacted by electric hydraulic press to produce a disc samples with dimension of 12.7 mm diameter and 8 mm thickness. The compacting pressure was 620 MPa for 1 minute. Graphite was used as a lubricant to reduce the friction during the pressing process.

The green compacted samples have been sintered in a vacuum tube furnace. The sintering system composed from the following parts: Tube furnace, Vacuum rotary, Quartz tube. The compacted samples were put in the quartz tube inside the tube furnace the samples were heated from room temperature to 850°C under vacuum condition. The heating rate was 30°C/min and the samples were kept for 6 hours at 850°C and then slowly cooled inside the furnace with preceding the vacuum circumstance at 10-3 torr. The prepared NiTi alloy converted to the powder form by crushing with ball mill of ceramic balls in planetary automatic ball mill (NϕM-O-O4). Then this powder was sieved with a 200 mesh sieve its normal aperture is 75 μm according to British standard-410.

Characterization: The characterization of prepared specimens was done by:

X - Ray Diffraction: The X - ray diffraction analysis was performed on amalgam powder, base Galloy, NiTi powder alloy and 5% NiTi/Galloy with X-ray machine (Shimadzu LabXXRD-6000, Japan) with copper Kα radiation at $\lambda = 1.5406$ A° and a nickel filter. The range of the diffraction angle 2θ° was (20-90°).

Scanning Electron Microscopy (SEM): SEM with high energy beam generated a verity of singles at the surface of solid specimen was used. This test was carried out using SEM type MIRA3 TESCAN/Czech Republic. This test was done for base alloy and 5% NiTi/Galloy.

Mechanical Properties

Hardness Test: Vickers microhardness test was done by a digital microhardness tester (Type TH715, Beijing, Time High Technology Ltd) with a static load of 200 g for 10 seconds at two different time intervals (1 day) and (7 days) after the trituration. Three recorders were measured at diagonal distribution across the specimen. The dimensions of the indentation are measured by the measuring microscope at 200× magnification.

Compressive Strength: The compressive strength was carried out using Instron machine, type Universal testing machine/WDW 200, China with the speed of 0.5 mm/min. The specimen was vertically put between the jaws. The test was done after (1 day) and (7 days) after the trituration at temperature of 37±1 °C. The compressive strength is calculated using the following formula:

$$\text{Comressive Strength} = \frac{\text{Max force}}{\text{Cross sectional area}} \quad \text{...(1)}$$

Diametral Tensile Strength: This test was done by the same machine of compression test at loading rate of (0.5 mm/min) after put the specimen at the lateral side and after (1 day) and (7 days) of trituration. The tensile strength was calculated by the following formula [13]:

$$\sigma_t = \frac{2P}{\pi DL} \quad \text{... (2)}$$

where: σ_t is a tensile strength (MPa), P is a load at fracture (N), D is a diameter of specimen (mm), and L is a length of the specimen (mm).

Creep: This test was done at 37±1 °C after (2 hours) and (45 min.) of trituration. And then the specimen was subjected to a constant axial pressure of 10 MPa and maintained for 21 hours. After that the shortening was obtained after measuring the length with micrometer caliper to calculate the percentage of creep according to A.D.A. Specification No.1 which allows the maximum of 3% creep [12]. The creep percent is calculated by the following formula [12]:

$$Creep\ (\%) = \frac{L_o - L}{L_o} \times 100 \qquad \text{...(3)}$$

where: L_o is original length (mm) and L is final length (mm).

Results and Discussion

Characterization: Fig. (1) shows the XRD pattern of powder alloy which identify the phase exist in the base alloy. The Cu_3Sn phase which has peaks at 2θ (41.787, 43.472 and 76.807) and the Ag_3Sn phase at 2θ (34.656, 37.602, 39.515, 39.589 and 52.059).

Fig. (2) shows the XRD pattern for base alloy (Galloy) where the formed phase are β-Sn, $CuGa_2$, $Ag_{72}Ga_{28}$, In_4Ag_9, Ag_3Sn and Cu_3Sn. The peaks of β-Sn are located at 2θ (30.644, 32.018 and 44.902), the peaks of $CuGa_2$ are located at 2θ (35.235, 44.576, 45.305, and 67.030), also the peak of In_4Ag_9 are located at 2θ (38.455) and for $Ag_{72}Ga_{28}$ are located at 2θ (38.922, 40.189, 41.245 and 57.20) and for Ag_3Sn phase are located at 2θ (34.656, 37.602, 39.515, 39.589 and 52.059).

Figure (3) shows the XRD pattern for NiTi alloy powder it can be observed the sintering temperature and time are enough to complete the phase transformation and all Ni and Ti transformed into monoclinic NiTi phase (M- NiTi), cubic NiTi phase (A-NiTi) and hexagonal Ni_3Ti phase).

Figure (4) shows the XRD pattern of 5% NiTi/Galloy, where the phase in this alloy is the same as base alloy except the addition of NiTi peaks that are located at 2θ (42.803, 46.534, 61.982 and 78.153) and the essential phases in base alloy.

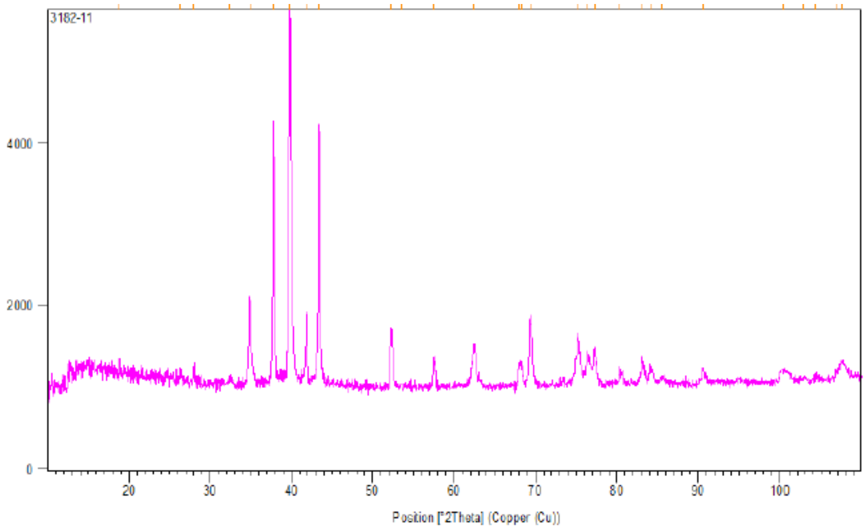

Fig. (1): XRD analysis of powder phase in Galloy.

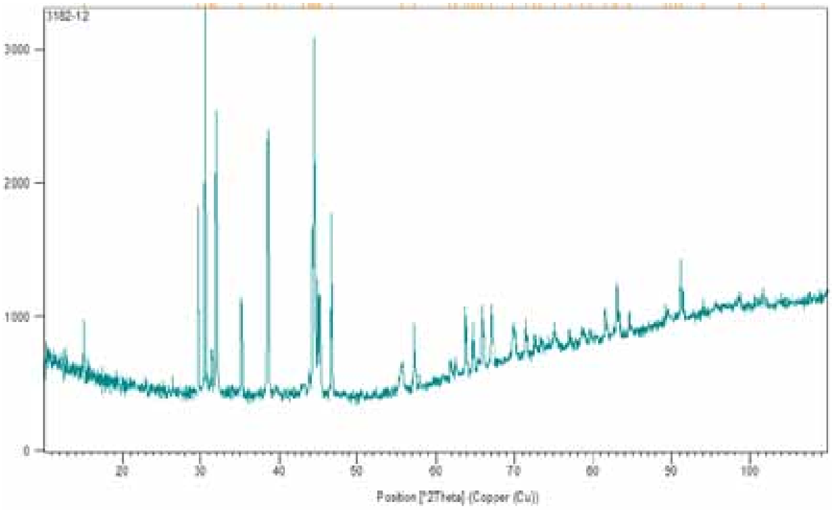

Fig. (2): XRD analysis of base Galloy.

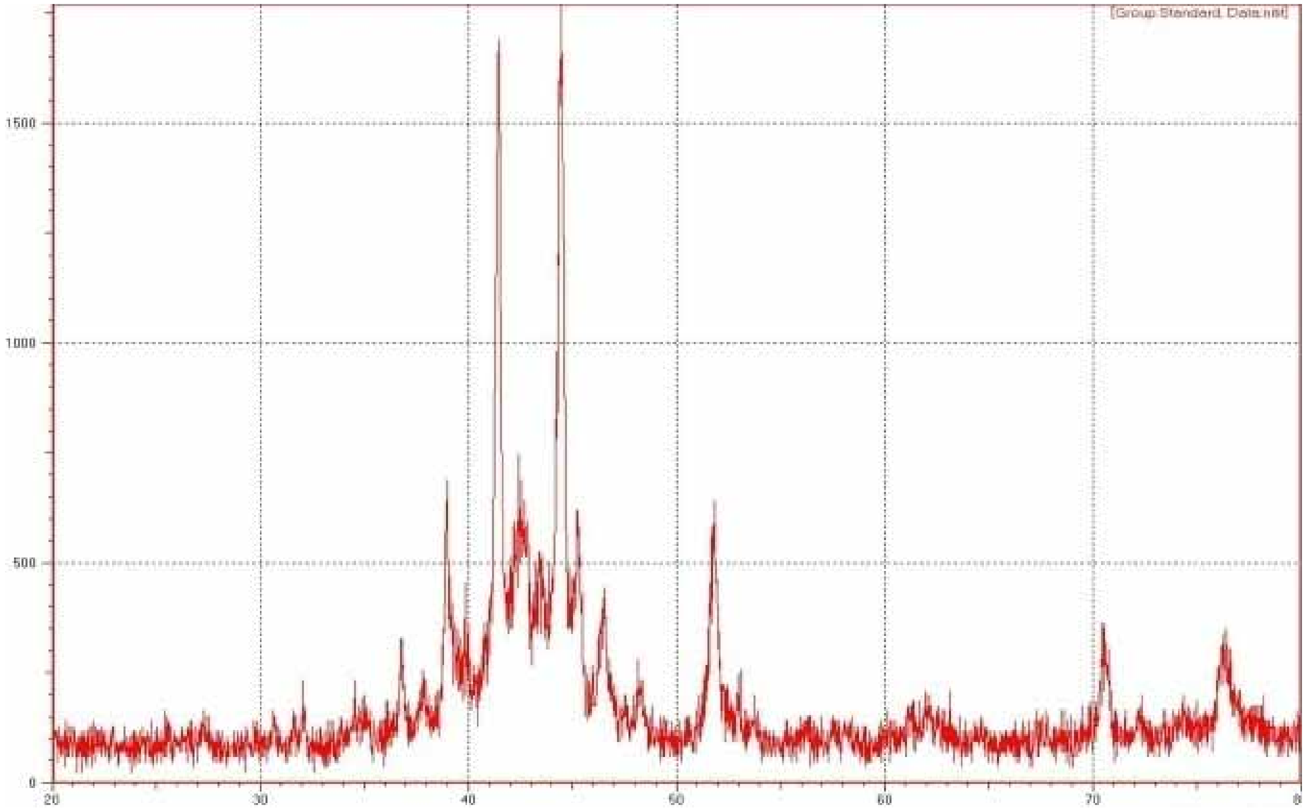

Fig. (3): XRD analysis of NiTi alloy powder.

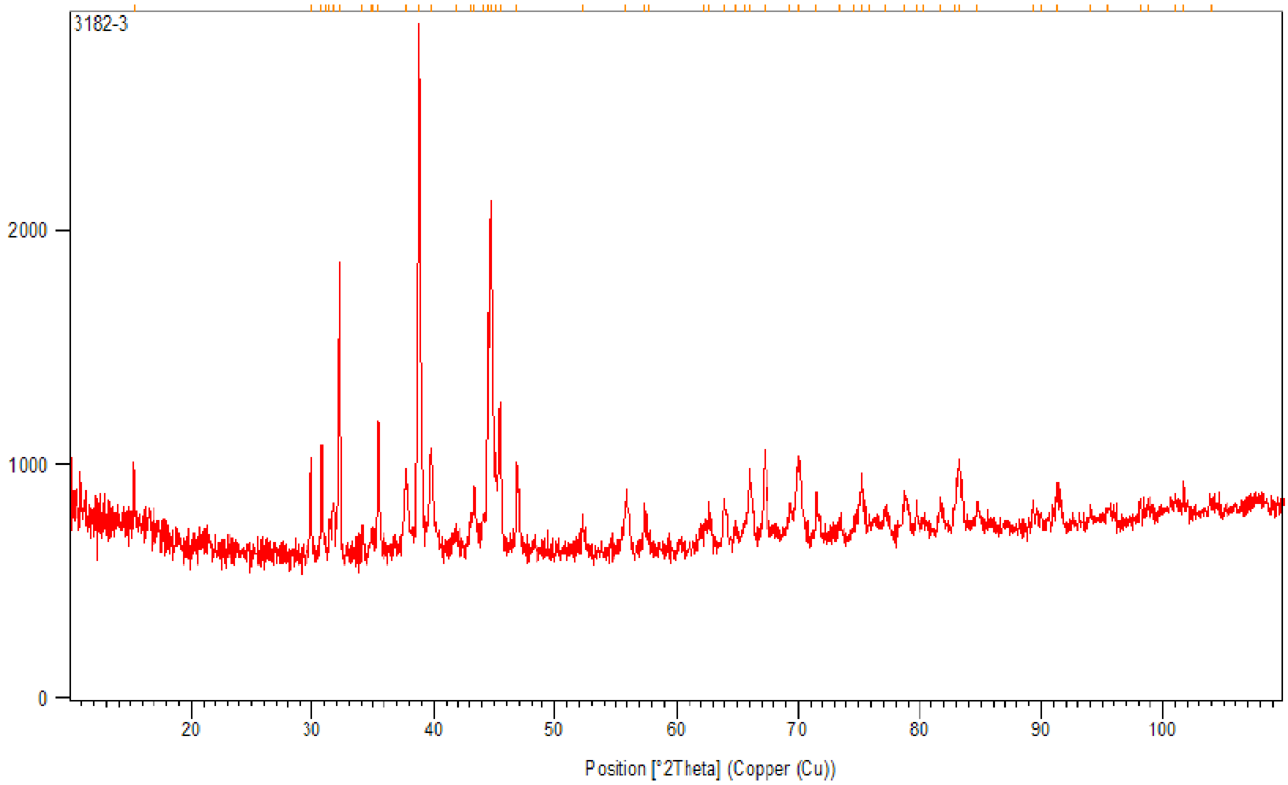

Fig. (4): XRD analysis of 5% NiTi/Galloy.

The microstructure of the base alloy is observed in the Fig. (5), the figure shows the unreacted cores of powder particles which consist of Cu + Sn + Ag surrounded by embedded zone within a complex matrix to produce $CuGa_2$. The core and the surrounding area denoted as A and C respectively and corresponding to the EDS test of the both region. The region B is referring to the matrix where it is consist of β-Sn and Ag_2Ga and Ag_9In_4, and the formed β-Sn is due to the following equation:

$$Cu_3Sn + 6\,Ga \rightarrow 3\,CuGa_2 + \beta\text{-}Sn \qquad ...(4)$$

While the D region is consist of $CuGa_2$phase, all the regions (A, B, C and D) are analyzed by EDS (Fig. 6) which indicates the presented elements and their distribution in base alloy.

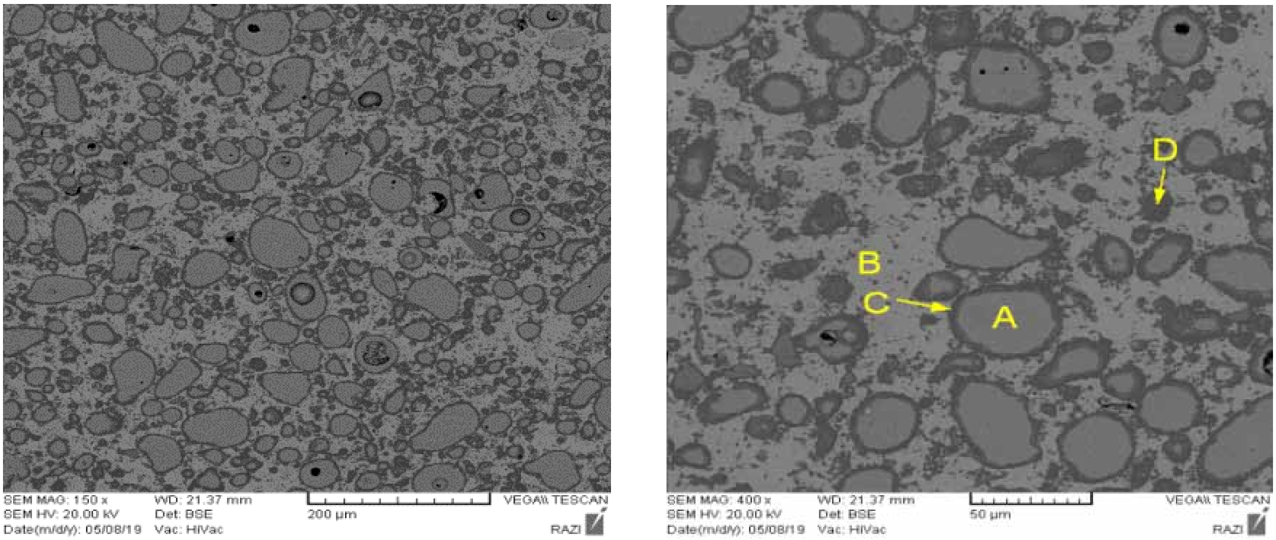

Fig. (5): SEM of base alloy (Ga alloy).

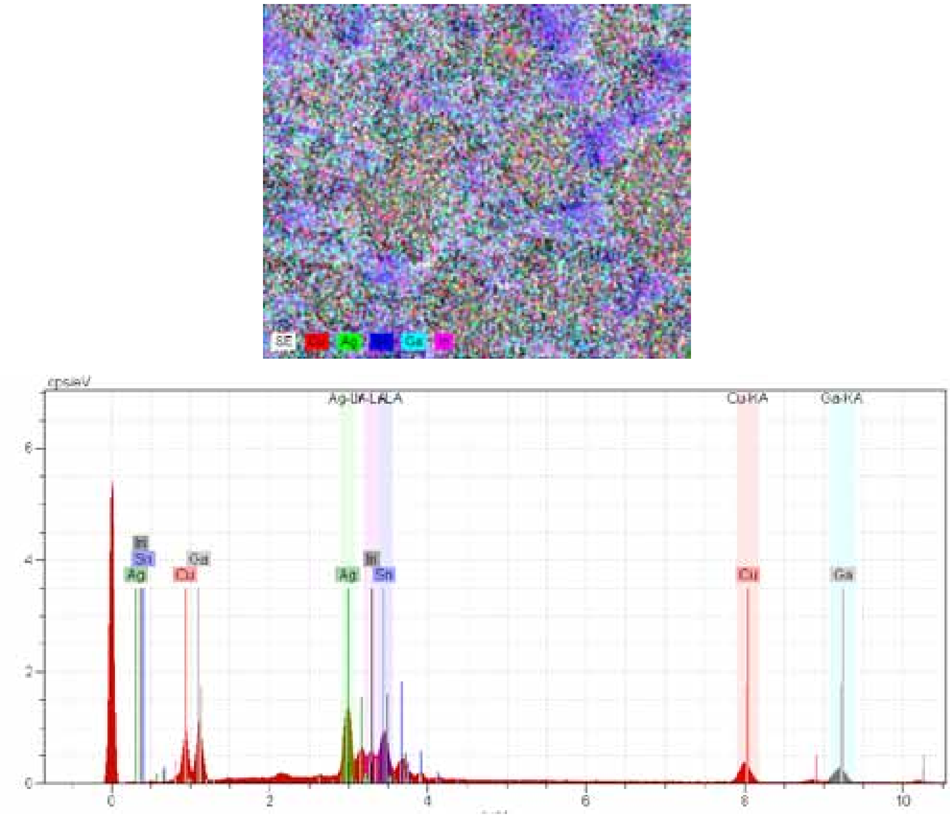

Fig. (6): EDS of base alloy (Ga alloy).

SEM of 5% NiTi/Galloy which indicates the main phases that presented in the base alloy. The phases are (β-Sn, $CuGa_2$, $Ag_{72}Ga_{28}$, In_4Ag_9, Ag_3Sn and Cu_3Sn) where the A region is consist of unreacted powders particles (Sn-Cu-Ag) surrounded by a reaction zone consist of β-Sn and $CuGa_2$ phase. While the B region is consist of NiTi alloy which confirmed by EDS analysis. The C region in SEM image is consisting of Cu, Ag, Ga, Sn elements and it may be $CuGa_2$or AgGa phase. The D region is consisting of β-Sn which is the result of the reaction that is mention in eq. (1). EDS analysis confirms the presence of NiTi within base alloy phases with uniform distribution and no sign to aggregation.

Microhardness: The microhardness test has been done for prepared specimens of Galloy with addition of different percentages of NiTi powder. The microhardness was determined after 1 and 7 days. The behavior shown in this figure indicates that the microhardness increased with increasing weight percents of additive compared with the base alloy without addition. The data of microhardness indicate that the hardness increased after 7 days compared with the values after 1 day because of the setting reaction doesn't complete immediately and some liquid alloy found and that will weaken the alloy, so after 7 days the setting reaction is complete and the alloy reach to its final strength. The increment of microhardness in presence of NiTi alloy powder is attributed to the influence of NiTi alloy nano powder to the Galloy which it is works through the filling of vacancies in the base alloy and also due to the unique mechanical properties of NiTi powder alloy that inhibit the dislocation movement.

Compressive Strength: The strength of a dental filling restoration must be high enough to resist the biting force especially compressive force. The produced specimens were tested after (1 day) and (7 days) and there is a high increase in the compressive strength after 7 days in the comparison with 1 day because the setting reaction is not complete at 1 day. The role of NiTi nanoparticles powder to increase the compressive refer to disoperation in the matrix and work to prevent the dislocation movement, and also the NiTi shape memory alloy with unique properties which have super elasticity also work to increase the compressive strength, and the good interfacial bounding between NiTinano powder and the base alloy.

Dimetral Tensile Strength: The Dimetral Tensile strength was done after (1 day) and (7 day) of trituration. the addition of NiTinano powder which led to increase the tensile strength with increasing of the NiTinanopowder alloy percentage, and the tensile strength increase after 7 days more than the increasing after 1 day, because the setting reaction dose not complete after 1 day.

Creep Test: The creep for NiTi/Ga alloys with different percentages (1, 2, 3, 4, and 5 wt%). The addition of NiTinanopowder alloy disperses in the matrix of the alloy and prevents the dislocation movement; also the NiTi powder particles have unique properties of super elasticity that work to increase the creep strength.

Conclusion

The addition of NiTi alloy powder to Galloy led to improve some mechanical properties of this alloy through the filling of vacancies in the base alloy and also due to the unique mechanical properties of NiTi powder alloy that inhibit the dislocation movement, this led to increasing the hardness, compression and tensile strength in addition to reduce the creep percent.

Financial Disclosure: There is no financial disclosure.

Conflict of Interest: None to declare.

Ethical Clearance: All experimental protocols were approved under the Department of Materials Engineering – University of Technology, Iraq and all experiments were carried out in accordance with approved guidelines.

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