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AMALGAM DENTAL FILLINGS AS THE SOURCE OF MERCURY EXPOSURE AND ALTERNATIVE SOLUTIONS

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ABSTRACT

The amalgam filling plays a major role in restorative dentistry when applied and has additional strength due to its contents, such as mercury and other alloys that could be Cooper, Silver, Tin, or Zinc. A concern has been raised that amalgam causes mercury toxicity and two primary organs are the kidney and the central nervous system that are affected by Hg poisoning.The objective of the present study is to examine new dental amalgam fillings and old amalgam fillings removed from patıent's teeth in term of a potential cause of mercury exposure, in addition to the alternative dental fillings. The technique of this study is a metallographic evaluation by using the rapid analytical technique of X-ray diffraction (XRD). Moreover, the specimens used have been obtained from a different set of amalgam fillings, to measure the mercury present in an unmixed sample, a mixed sample according to the manufacturer's recommendations, and dental amalgam fillings that had been in the patient's theeth after filling them for 15 and 20 years to analyze exposure to mercury by comparison.
Mercury unmixed sample contains on silver Tin 64% and Copper Zinc 36% While mixed samples contain of Silver Mercury 63% and Copper Tin 37%, the sample after 15 years' dental amalgam filling induced when the Silver mercury contain is increased to 84% and low Copper Tin 16%, While the sample after 20 years' dental amalgam filling containing approximately Silver Mercury contain is 80% and Copper Tin 20%.

1. INTRODUCTION

Liquid Mercury and an alloy are the two primary components of the vast majority of Mercury-containing fillings, which is a metal powder. When the word alloy is used in Physics, it describes a mixture of one or more elements, at least one of which is a metal. Briefly, the term "amalgams" refers to mixes of mercury and one or more other metals (such as Silver or Copper), which may be dissolved into the Mercury or which may just be metal particles that the Mercury has bonded together. The maker of dental amalgam sets the alloy/Mercury mixing ratio at a level where it is purported that the Mercury is firmly linked to the alloy [1]. In developing nations, silver amalgam is still the material of choice for dental treatment because of its superior mechanical qualities, low cost, and ease of insertion. More than 100 million Silver-amalgam fillings are reportedly inserted into American mouths each year, according to the American Dental Association (ADA) [2]. Conversely, according to Mason [3]. Inorganic mercury (in the form of Mercury salts), metal Mercury (also known as elemental Mercury), and organic Mercury are all naturally occurring forms of the hazardous metal Mercury (Hg) in the environment (methyl Mercury being the most common). Dental amalgam is a Mercury-based filling composed primarily of Silver (35%) and trace amounts of Tin (9%), Copper (6%) and Zinc (3%); it has a metallic mercury content of about 50% (Hg0). Although a little quantity of Mercury in the form of Hg0 is continuously released from surface of an amalgam fillings into the mouth's saliva and mouth air or grinding motions. The two primary organs are the kidney and the central nervous system that are affected by Hg poisoning [4]. Additionally, according to report of Vimy [5], estimates of the typical amount of mercury emitted by dental amalgams varies from 3 to 17 mg/day depending on the total number of amalgam fillings. The majority of researches [1-6] have demonstrated that amalgam fillings gradually leak Mercury vapor, with the lungs being the primary route via wherein 80% of the Hg0 that is breathed in enters the blood. As an illustration, the Ministry of Foreign Affairs of Norway of

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Environment passed this law banning the manufacture, import, export, sale, and use of substances containing mercury [7]. The method dentists perform operative dentistry has significantly changed in recent years, and one such shift is the increased use of resin composites for the repair of posterior false teeth [8]. The improvement of resin composite materials, bonding methods, and processes, greater patient demand for more cosmetic restorations over Silver amalgam, and increased predictability of resin composite performance in posterior teeth have all contributed to this [9, 10].

The usage of resin composite in occlusal and occlusal proximal cavities as a direct restorative material is currently supported by data in the dental literature [10]. Posterior resin composite restorations put properly may be just as functional as those made with Silver amalgams [9]. Resin composite offers a more aesthetic appearance than Silver amalgam [11], eliminates the need to remove healthy dental tissue for retention, lowering the danger of eventual tooth fracture, and strengthens the remaining tooth material [12]. Resin composites have more technique sensitivity, take longer to place a posterior replacement, and are more expensive than Silver amalgam amalgams [9-13].

Based on the previous show and because the prescribed limits for daily Mercury intake are frequently reached or exceeded by the Mercury released over time from a few amalgam fillings. Hence, Mercury from corroding amalgam fillings offers a potential health hazard. So, the main goal of the current study was focused on this phenomenon, which is the emission of metallic Mercury (Hg0) from the surface of the fillings due to the formation of droplets rich in Mercury on the surface of the amalgam filling, as result of corrosion, grinding motions, chewing.

Also, we can estimates released Mercury amounts by measuring the depth and type of corrosion attack. In this study metallographic evaluation was done by using the energy dispersive x-ray technique. Moreover, the specimens used have been obtained from a different set of amalgam fillings used on technique was used to analyze exposure to mercury present in an unmixed sample, a mixed sample according to the manufacturer's recommendations, and dental amalgam fillings that had been in the patient's mouth after filling them for 15 and 20 years.

2. MATERIAL AND METHOD

This work aims to study metallographic evaluation was done by using the Energy Dispersive X-Ray Technique. Moreover, the specimens used have been obtained from a different set of amalgam fillings used on technique was used to analyze exposure to Mercury present in an unmixed sample, a mixed sample according to the manufacturer's recommendations, and dental amalgam fillings that had been in the patient's mouth after filling them for 15 and 20 years.

Amalgam was selected in this study because it is used extensively for direct restoration that involves filling a cavity in the tooth structure in order to return the tooth's form and function to normal. To eliminate dental caries, this cavity is created inside the tooth.

Amalgam dental filling material manufactured by (Crown Alloy) Turkey; the material is provided in the form of Amalgam Sample mixed together by the amalgamator. The main constituent of Dental amalgam is an alloy created when Mercury (Hg) and Silver-Tin combine (Ag-Sn). Additionally, there are fluctuating concentrations of Copper (Cu) and trace levels of zinc (Zn). A combination of Ag, Sn, and Cu makes up the remaining portion of the final filling material after the Mercury (Hg) and powder alloy react.

In this study other materials, instruments and equipment were used in different steps of the sample's construction and testing.

The first step of the experimental work for samples preparation and testing for the amalgam dental filling was done in dental clinic and the other steps of measurements of x-ray diffraction were done in dental materials laboratory.

2.1. Samples Preparation

These amalgam dental filling materials were the specimens used have been obtained from a different set of amalgam fillings used on technique was used to analyse exposure to mercury present in an unmixed sample, a mixed sample according to the manufacturer's recommendations, and dental amalgam fillings that had been in the patient's mouth after filling them for 15 and 20 years.

2.1.1. Unmixed Sample

Amalgam dental filling material manufactured by (Crown Alloy) Türkiye; samples were fabricated as the following instructions: The plate is made of metal specifically according to the sample size that required by the XRD device. Take a capsule of the amalgam manufactured by the company and open it on plate metal to prepare it for steps of measurements of x-ray diffraction.

2.1.2. Mixed Sample

Amalgam dental filling material manufactured by (Crown Alloy) Türkiye; samples were fabricated as the following instructions: 20 amalgam fillings mixed by the amalgamator device for 10 seconds and placed one by one in the sample metal form, smoothed and left two hours to dry and firm. Then the sample is analysed by XRD device according to manufactures' instructions and recommendations were had done in (EL Masah dental clinic, Tajoura, Libya).

2.1.3. Old Samples 15 and 20 Years

Dental amalgam fillings that had been in the patient's mouth after has been filled for 15 and 20 years. This step in the clinic, which included amalgam fillings, was for my patients in the clinic Thus; ethical approval was obtained from the patients from whom the amalgam fillings were taken after they were removed from the patient's mouth. Dental amalgam fillings that had been in the patient's mouth after filling for 15 and 20 years were ground and filled into a metal plate to prepare them for the steps of X-ray diffraction measurements.

2.2. X-Ray Diffraction Analysis

An analytical method based on the diffusion of X-ray radiation by a substance is called X-ray scattering. X-ray scattering can be utilized for crystalline or amorphous materials, whereas X-ray diffraction could only be employed with crystalline materials. The basis for X-ray scattering is the interaction of X-rays with atomic electrons. For materials with dimensions under one micrometer, such as big molecules (such as proteins and polymers) and nanoparticles (such as nanotubes), X-ray scattering can reveal their shape, size, and orientation. The substances that are evaluated can be foams, gels, liquids, solids, and more. Because this method is non-destructive, it can be applied to delicate materials, the XRD machine is displayed. The XRD test was done (carried out at the laboratories of the Libyan petroleum institute, Tripoli, Libya). The data collected and the mean of four amalgam dental filling samples for the all-samples amalgam materials was measured, calculated and analyzed using XRD.

3. RESULTS AND DISCUSSION

3.1. X-Ray Diffraction Data

In this section the experimental result will discussion X-ray diffraction data analysis of amalgam fillings for mercury exposure was performed by the sol-gel method using X-ray diffraction (XRD). Amalgam dental fillings materials were analyzed with a PW 1800 X-ray Diffractometer, XRD patterns were recorded in the temperature range: 26.4 °C Relative humidity: 48%RH. The intensity of the obtained diffraction peak was determined by comparison with standard data having theta angle with higher intensity using JCPDS: 075-1687. The manufacturer of amalgam dental fillings, the peak intensity was mercury exposure. X-ray diffraction (XRD) technique was used to analyze exposure to mercury present in an unmixed sample, a mixed sample according to the manufacturer's recommendations, and dental amalgam fillings that had been in the patient's mouth after filling them for 15 and 20 years.

3.2. Unmixed Sample Dental Amalgam Filling

The results are shown unmixed sample in the Figure 1 revealed two main phases for the measured amalgams: Ag3Sn and Cu4Zn in the XRD patterns were recorded from 2θ range of 26.4 °C.

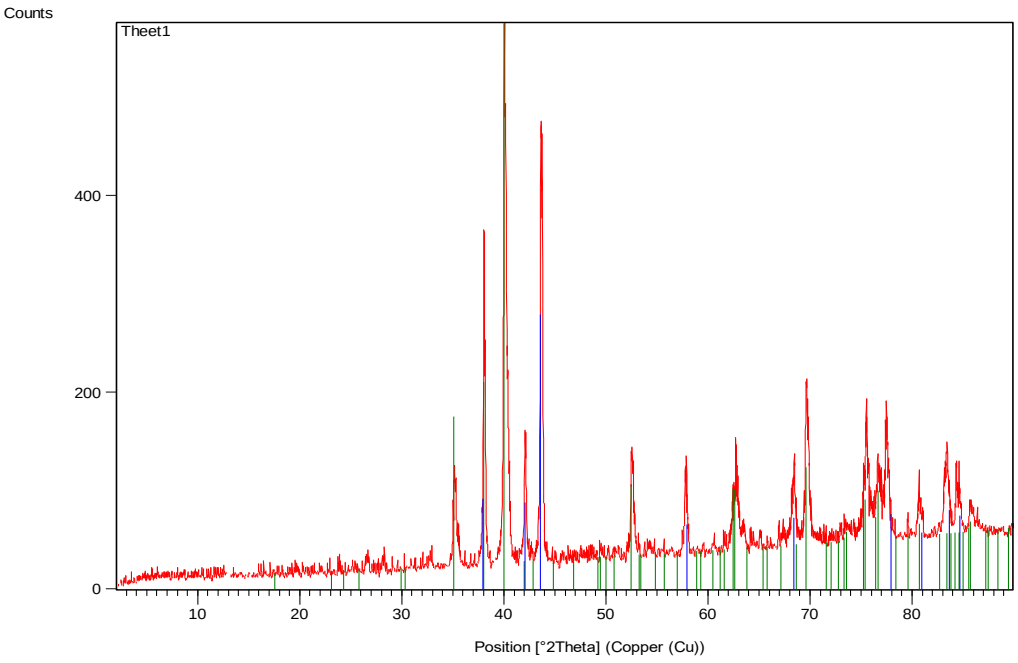

Fig. 1. Analyze view of unmixed sample dental amalgam filling

In Table 1, there is analyze view of unmixed sample in the amount of Mercury available in pattern list & semi quantitative dental amalgam filling company unmixed was Ag3Sn 64 %, Cu4Zn 36%.

Table 1. Analyze view of unmixed sample dental amalgam filling

Ref. Code	Compound Name	Chemical Formula	Semi quantitative %
01-071-0530	Silver Tin	Ag_3 Sn	64
03-065-6066	Copper Zinc	Cu_4 Zn	36

3.3. Mixed Sample Dental Amalgam Filling

The results are shown mixed sample in the Figure 2 revealed two main phases for the measured amalgams: Cu10Sn3 and Ag2Hg3 in the XRD patterns were recorded from 2θ range of 26.4 °C.

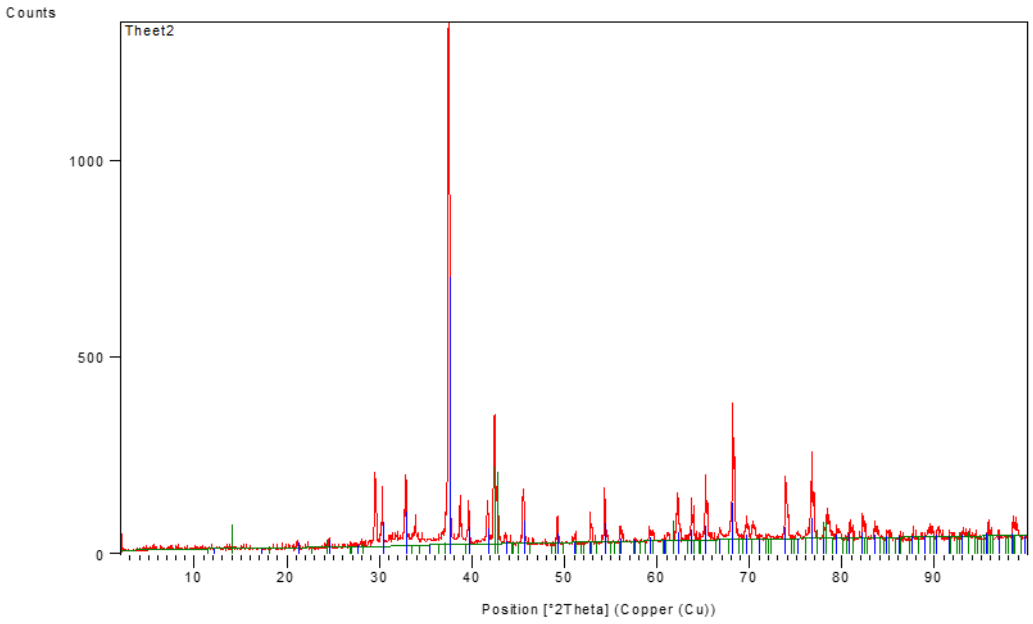

Fig. 2. Analyze view of mixed sample dental amalgam filling

In Table 2, there is analyze view of mixed sample in the amount of mercury available in pattern list & semi quantitative dental amalgam filling company mixed was Cu10Sn3(37%), Ag2Hg3(63%).

Table 2. Analyze view of mixed sample dental amalgam filling

Ref. Code	Compound Name	Chemical Formula	Semi Quantitative %
03-065-3632	Copper Tin	$Cu_{10}Sn_3$	37%
03-065-6103	Silver Mercury	Ag_2Hg_3	63%

3.4. Sample After 15 Years Dental Amalgam Filling

The results are shown after 15 years dental amalgam filling in the Figure 3 revealed two main phases for the measured amalgams: CuZn and Ag2Hg3 in the XRD patterns were recorded from 2θ range of 26.4 °C.

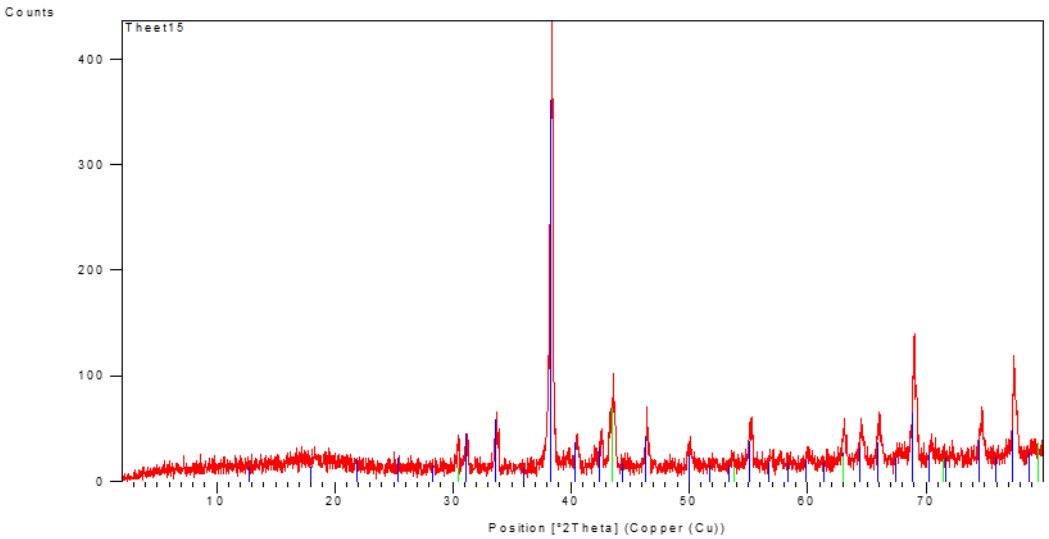

Fig. 3. Analyze view of sample after 15 years dental amalgam filling

In Table 3, there is analyze view of samples after 15 years in the amount of Mercury available in pattern list & semi quantitative dental amalgam filling company mixed was CuZn (16%), Ag2Hg3(84%).

Table 3. Analyze view of sample after 15 years dental amalgam filling

Ref. Code	Compound Name	Chemical Formula	Semi quantitative %
03-065-6103	Silver Mercury	Ag_2Hg_3	84
03-065-9061	Copper Zinc	CuZn	16

3.5. Sample After 20 Years Dental Amalgam Filling

The results are shown after 20 years dental amalgam filling in the Figure 4 revealed two main phases for the measured amalgams: CuZn and Ag2Hg3 in the XRD patterns were recorded from 2θ range of 26.4 °C.

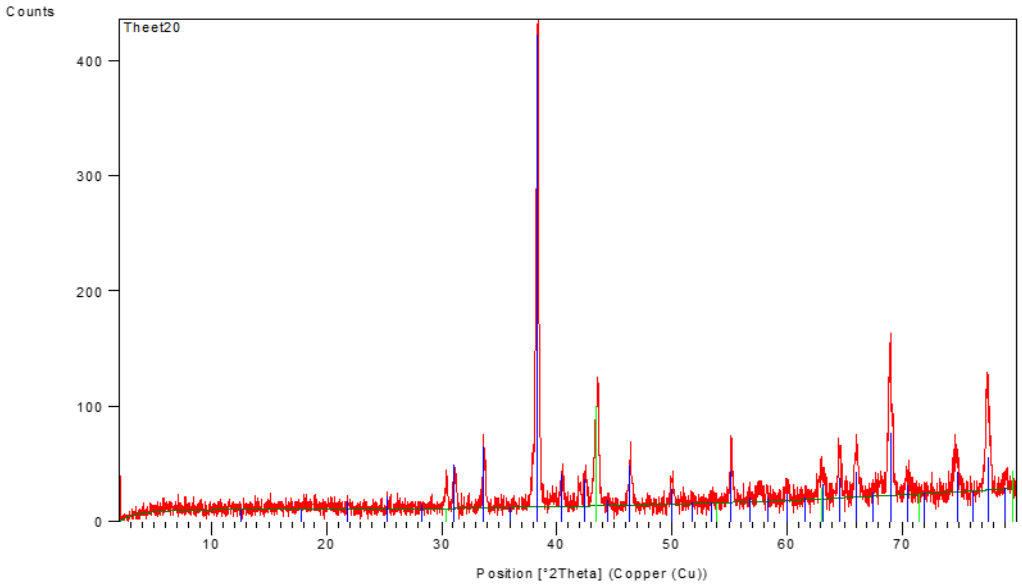

Fig. 4. Analyze view of sample after 20 years dental amalgam filling

In Table 4, there is analyze view of samples after 20 years in the amount of Mercury available in pattern list & semi quantitative dental amalgam filling company mixed was CuZn (20%), Ag_2Hg_3(80%).

Table 4. Analyze view of sample after 20 years dental amalgam filling

Ref. Code	Compound Name	Chemical Formula	Semi quantitative %
03-065-6103	Silver Mercury	Ag_2Hg_3	80
03-065-9061	Copper Zinc	CuZn	20

This was an in vivo study which involves the use of remove dental amalgam filling human. Therefore, consent was obtained from the patients.

In the present study the amount of mercury released from dental amalgams with silver content unmixed sample, a mixed sample according to the manufacturer's recommendations, and dental amalgam fillings that had been in the patient's mouth after filling them for 15 and 20 years.

The results of this study showed there is no mercury unmixed sample. This sample contains on silver Tin 64% and copper zinc 36% while mixed samples contain of silver mercury 63% and copper Tin 37%, the sample after 15 years' dental amalgam filling induced when the silver mercury contain is increased to 84% and low copper Tin 16%, while the sample after 20 years' dental amalgam filling containing approximately silver mercury contain is 80% and copper Tin 20%.

The Silver-Mercury phase (1) is the dental amalgam structure's matrix, and it has a significant impact on its mechanical behavior and interaction with the environment. In addition, amalgam restorations are the primary source of Mercury emitted. A contains roughly 67-70 percent Mercury as part of the dental amalgam composition.

The second approach, X-ray diffraction (XRD), takes advantage of the fact that atoms in alloys form crystal lattices, allowing for the identification of specific phases in materials.

The goal of the study was to use XPS and XRD techniques to analyze the general composition of seven distinct commercially available high Copper dental amalgams (HCSS). We also looked into the chemical relationships between amalgam components and the oxygen reactivity of the key metallic elements in the amalgams under investigation.

MotohiroUo et al., according to estimates, the enhanced surface layer is only a few nanometers thick this basically corresponds to the XPS study's depth. Furthermore, their being metallic components such as Zinc and Tin, which quickly create a surface oxide layer that is permanent, inhibits mercury release [14].

E. Talik et al., within high Copper dental amalgams, the optimum quantities and ratios of fundamental elements Ag, Cu, and Sn play a vital influence in mercury release behaviors and chemical properties. The increase in mercury evaporation is influenced by a high Sn/Ag ratio (1) in some of the investigated amalgams. Copper and Silver concentrations affect the oxygenation reactivity of metallic elements in dental amalgams. The findings of this study corroborate previous findings. The corrosion resistance of dental amalgams is affected by the relative quantities of phases. The presence of pure Hg in the amalgams may imply that, as previously mentioned, the triturating circumstances can impact the release of this element [15].

Mahmoud Bahari Reported the Silver content of dental amalgam is inversely proportional to the amount of mercury released [16].

Ulf G. Bengtsson showed the non-ɣ2-amalgams are touted as being stronger and more corrosion resistant. A significant sub-optimization has happened when attempting to satisfy these development goals. These amalgams, which were developed in the 1970s, emit around ten times more mercury vapor in experimental setups than the ones previously employed. Ordinary dental staff, politicians, and other decision-makers are unaware of the instability of current non-ɣ2- amalgams [17].

The fractured samples' spectra revealed the presence of Carbon. The relative concentrations of the various elements are critical for the XPS investigations. Using the peak area and peak height sensitivity factors, the Multipack Physical Electronics application can quantify XPS spectra. The typical atomic concentration computation yields a ratio of each component to the total number of other components in the data set. Only those elements were taken into account for which a distinct line can be seen in the spectrum. For those lines, the backdrop was removed, the line's region's limit was individually picked, and then integration was performed [18].

The association between the makeup and the mercury release rate of several type of dental amalgam was investigated. In the g1 phase, mercury emissions reduced as the tin level increased. Zinc, on the other hand, did not have the same effect as Tin. The key predictor of Mercury vapor release, according to this paper, is the tin level in the g1 phase. The creation of a surface oxide layer composed of Tin and Zinc, as well as its effect on mercury leakage, were validated in an ongoing work and this investigation by Berglund et al., which is consistent with Ferracane et. al., on the other hand, had a greater mercury release than the other indium-free amalgams because its surface was rapidly coated by indium with oxygen. As a result, indium's surface layer was shown to be less effective than Tin and Zinc at preventing Mercury emission [19].

4. CONCLUSION

Within the limitations of the study, the following conclusions can be drawn:

•There are some changes that occurred in the unmixed sample dental amalgam filling in terms of compounds, as the Silver Tin compound and another Copper Zinc compound appeared. As for the Silver Mercury element, it did not appear in the results.

•Dental amalgam filling mixed in terms of compounds, where the Silver Mercury compound appeared with a high value before it was filled inside the dental cavity and the other Copper Zinc compound appeared.

•A sample after dental amalgam filling for 15 and 20 years in terms of compounds, where the Silver Mercury compound appeared in a higher value than the filling of the mixed sample and the other Copper compound appeared.
•Unmixed sample and mixed sample dental amalgam filling to new technological effect gives rise to decrease the Mercury in the amalgam.
•A sample after dental amalgam filling for 15 and 20 years there was no like process might be leakage.

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Competing interests
The authors have no competing interests to declare that are relevant to the content of this article.

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