Jurnal Teknologi

Full paper

Non-Destructive Assaying Gold Jewellery Using Dual-Energy Micro-Computed Tomography

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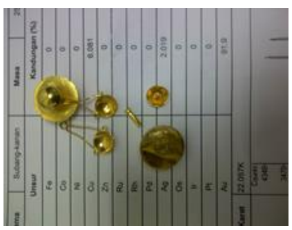
Article history

Received :15 August 2014
Received in revised form :
5 January 2015
Accepted :10 February 2015

Graphical abstract



Abstract

Determining the fineness of gold jewelleries remains one of the most challenging tasks in gold trading. The existing technology of gold testing is inadequate, allowing gold counterfeiting worldwide. The most popular non-destructive method for analysis of gold jewelleries is X-ray fluorescence technique. However, the technique is limited to surface only and it is also greatly influenced by matrix effects. In this paper, dual-energy X-ray micro-computed tomography method was proposed to assay gold jewelleries. Experimental results demonstrated that grey values of reconstructed tomographic images in combination with advanced image analysis procedures could be used to detect fake jewelleries. Due to the uniqueness of X-ray absorption, the technique was also capable of identifying different materials in gold jewelleries. Further analysis on sectioned-earrings samples using X-ray diffraction techniques and visual observation confirmed all tomographic findings.

Keywords: Non-destructive evaluation; gold purity; gold scam prevention; fake gold; X-ray fluorescence; dual-energy X-ray micro-computed tomography; image reconstruction

1.0 INTRODUCTION

Due to the high demand for gold and its accompanying high price, the jewellery market is flooded with tungsten or other metal articles plated with thin layers of gold purporting to be gold products, but instead are fakes. When purchasing a gold item, the purchaser typically evaluates the gold to determine its worth. This is usually a very fast process that does not permit detailed analysis. Purchasing gold-plated jewelleries when represented as a solid gold or solid gold alloy resulted in a significant loss from a purchase transaction.

Due to its high value, the analysis of gold jewelleries must be carried out with high precision and accuracy. The classical approach for testing the fineness of gold in a non-destructive manner, is the fluid immersion test, first proposed and used by Archimedes [1]. The so-called densimeter was designed and manufactured based on this principle. Applying such test to a fake gold product made of a tungsten core surrounded by a thin layer of gold plating would probably not give a definitive answer, since the density of gold (ρ_{Au}=19.3 gm/cm^3) and tungsten (ρ_W=19.25 gm/cm^3) are almost identical [2]. Furthermore, a densimeter cannot be used to test hollow jewelleries which may be found in various shapes and sizes. At present, fire assay (or cupellation) remains the best analytical test to find out the composition of elements in gold jewelleries. However, this procedure resulted in a sample being destroyed as it should be melted [3]. Nevertheless, the cupellation, owing to its high precision and accuracy, is still the preferred method for the determination of gold in jewellery products [4, 5]. Among nuclear techniques, the X-ray fluorescence (XRF) technique is the most popular for such non-destructive assaying of gold products [6]. Different XRF systems with different configurations are available, each designed for a specific purpose. However, XRF sometimes does not help in detecting fraud. The technology is limited to testing the surface (typically good for < 20 µm from the surface) and knowledge about the inner details of test samples remains unknown [7]. This work presents the use of a dual-energy X-ray micro-computed tomography imaging method to analyse fake gold earrings. Using this method, a volumetric measurement can be carried out without destroying the sample and at the same time, offering a non-contact measurement. The technology is being appraised by the Malaysian Nuclear Agency, Institute of Research and Consultancy YaPEIM (INPUT YaPEIM) and Fraunhofer EZRT, Germany in order to support the establishment of YaPEIM's Non-Destructive Assaying Centre (NDAC) for gold [8, 9]. This paper reports some of the preliminary results.

2.0 MATERIALS AND METHODS

2.1 Dual-Energy X-Ray Computed Tomography–Basic Principle

Computed tomography (CT) using an X-ray source is an imaging technique that can provide a two or three dimensional cross-sectional view of the interior of an object. The X-ray transmission data are collected by radiation detectors at many different angles within the image plane, and these data are then used to reconstruct a meaningful cross-sectional image that is essentially a map of the density distribution. The introduction of the CT scanner revolutionized the field of medical diagnostic imaging as it provided more detailed diagnostic information than the previous non-invasive imaging techniques. For the same reason, in the last two decades, the tomographic imaging method has been used increasingly for non-medical applications. The principle of X-ray CT imaging can be found in many literatures [10, 11].

The attenuation of X-rays in the CT energy range results predominantly from only two effects: Compton scattering and photoelectric absorption. A third effect, called Rayleigh scattering, is appreciable only at very low energies, clearly below 30 keV. Due to heavy pre-filtration the X-ray spectra used in CT system contains relatively few X-ray photon at energies below 30 keV and Rayleigh scattering can thus be ignored. Dual-energy CT methods have been developed for many years for medical application, but have lately became a topic of high interest in non-destructive evaluation of materials. Supposed two measurements are conducted at different X-ray energies, at a high energy E_H and at a low energy E_L, where I and I_0 represent the measured primary and attenuated intensity, respectively. Two equations containing the unknown parameters δ_1 and δ_2 represent the line integrals over the basis materials densities. The terms $(\mu/\rho)_1$ and $(\mu/\rho)_2$ are the mass attenuation coefficients of materials 1 and 2, respectively,

$$I_H = \int I_{0H}(E).\exp\left[-\left(\frac{\mu}{\rho}\right)_1 (E)\delta_1 - \left(\frac{\mu}{\rho}\right)_2 (E)\delta_2\right]dE \qquad (1)$$

$$I_L = \int I_{0L}(E).\exp\left[-\left(\frac{\mu}{\rho}\right)_1 (E)\delta_1 - \left(\frac{\mu}{\rho}\right)_2 (E)\delta_2\right]dE \qquad (2)$$

By solving equations (1) and (2), two unknowns can be determined. In the present work, materials 1 and 2 represent gold and non-gold, respectively. The solution process described above is based on directly using the measured data before image reconstruction. It is possible just the same to perform the basis material decomposition using reconstructed CT images.

2.2 Gold Earrings Sample

Counterfeiting of valuable goods or merchandise is well-known worldwide. Liquor, cigarettes, clothing, compact discs, paper money and other false abundant open market, especially in countries that do not have advanced tracking technology or policy enforcement and control is less strict. Similarly, counterfeiting gold and precious metals are also not a new thing. Gold earrings sample which is used in the experimental work was obtained from an Ar-Rahnu YaPEIM's pawnshop, and it is believed to be fake gold jewellery traded by an international syndicate. It is a semi-spherical shape with based of about 18 mm diameter, and 10 mm height including a small leg, as shown in Figure 1. The out-side shape is smooth and perfect with no significant dent on it, shiny and golden yellowish in color. It has been tested for purity and fineness by using conventional densimeter and XRF techniques but results were very doubtful and they did not match each other. It is suspected that the earrings are fake. Based on these findings, the earrings were taken as a test piece for this extensive evaluation work. No sample preparation was needed and the sample was analysed using an X-ray micro CT system, as received.

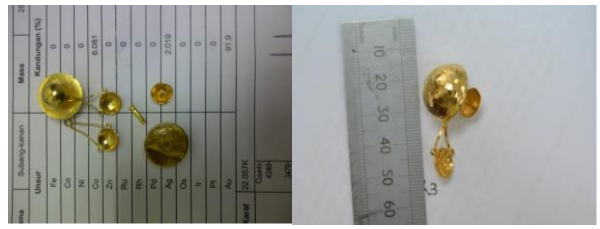


Figure 1 A pair of gold earrings (suspected fake) with associated accessories which was used in this CT imaging investigation. The sample was obtained from an Ar-Rahnu YaPEIM pawnshop in Kuala Lumpur

2.3 X-Ray CT – System and Method

The experimental work was conducted at the Fraunhofer EZRT in Furth, Germany, using a high-resolution X-ray micro-computed tomography system. The key components of the X-ray CT system are represented in Figure 2. The earrings sample was placed on a high precision turntable; the source and the detector were fixed, while the sample was rotated during measurement. A unipolar microfocus X-ray tube with adjustable voltage from 20 to 240 kV and tube current 50 to 300 µm was used. For the purpose of dual-energy measurement approach, X-ray operating voltage settings of 140 kV and 225 kV were used to represent lower and high energies, respectively. A flat panel X-ray detector, Varian 4343CB with 3048 x 3048 pixels was used to record the transmission of the conical X-ray beam through the sample. The distance source–object–detector was adjusted to produce images with a pixel size of 10 µm. Four frame averaging, a rotation step of 0.50^o and an exposure time of 1475 ms were chosen to minimize the noise, covering a view of 360^o rotation. Smoothing and beam-hardening correction steps were applied to suppress noise and beam hardening artifacts, respectively. Beam hardening correction was only moderately applied due to the use of aluminum or copper filter during acquisition. Once initial parameters were set, the acquisition step was completely automated. Setting and scan time, on average, required 20 min.

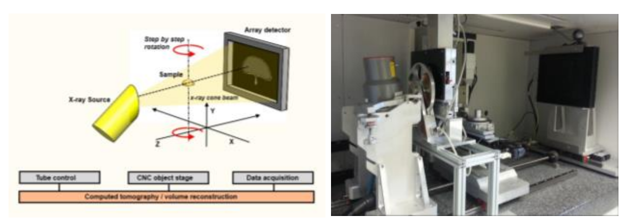


Figure 2. A schematic diagram (left) and a photograph (right) of a cone beam X-ray micro computed tomography system using a flat panel detector and a cone beam X-ray source.

A fast ring artifacts reduction was applied during image reconstruction processes. A set of flat cross-section images, was obtained for the sample after tomographical reconstruction by the reconstruction software, Fraunhofer's Volex 6. Three-dimensional reconstructions of samples were created by effectively stacking all two-dimensional tomographs. This was performed by using Fraunhofer's 2X Processing Suite and Visualisation software, VolumePlayer 6.6. Further analysis and adaptive rendering on the reconstructed images was carried out by using commercially available VolumeGraphic (VG Studio) Max 2.2.5 software.

■3.0 RESULTS AND DISCUSSION

3.1 Dual-Energy X-Ray CT Image Analysis

Figure 3(a) to 3(d) show the reconstructed cross-section images (three 2D and one 3D images) of the earrings acquired by X-ray CT using the dual-energy approach (140 kV and 225 kV). The grey contrasts in these images are based on the differences in absorption of X-rays by the constituents of the sample (e.g. gold, silver and air). This contrast is produced by a variation of density and a change in composition of the sample and is based exclusively on the detection of an amplitude variation of X-rays transmitted through the sample itself. The obtained image is a map of the spatial distribution of the X-rays in which the brighter regions correspond to the higher level of attenuation, i.e. higher density region (gold). It can be assumed from these images that the darkest areas represent the voids (air gaps) as it has a lower absorption coefficient with respect to other structures. Clearly, all three 2D images in Figure 3(a) to 3(c) show that the earrings constitute of three different materials. First, the outer layer which corresponds to the gold layer, second the air gaps and third the inner part which consists of a quite uniform density material. Using image analysis software VolumePlayer 6.6, the thickness of the gold layer is measured to be about 300 µm. The air gaps are not uniformly distributed below the gold layer where at some points the gap is large, particularly at corners between the flat base and the spherical shape, whereas the others no gaps exist. The inner part is obviously not the same material as the outer layer, as can be seen by large different in the grey level. Thus, the inner part is not gold.

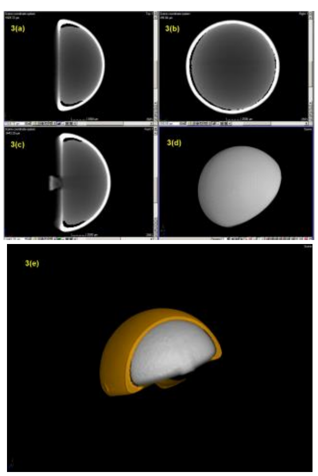


Figure 3 Reconstructed X-ray computed tomography images (2D and 3D) of a fake gold ear-ring using dual energy approach of 140 kV and 225 kV. The images revealed that the ear ring is made of two different metals (outer layer of about 288-300µm thick is 22K gold and inner part is silver), and air voids are clearly be seen in certain locations between these two precious metals

Figure 3(e) demonstrates a 3D pseudo colour plot of the same X-ray CT data using the VGStudio Max 2.2.5 software. As can be seen very clearly, three different materials are used to manufacture the earrings. The outer layer and the inner portion are 22K gold (ρ_{Au}=17.45 to 18.24 g/cm^3) and silver (ρ_{Ag}=10.49 g/cm^3), respectively. Again, the air gaps are visible at the areas between the flat base and the spherical portion.

3.1 XRF and SEM-EDX Analysis

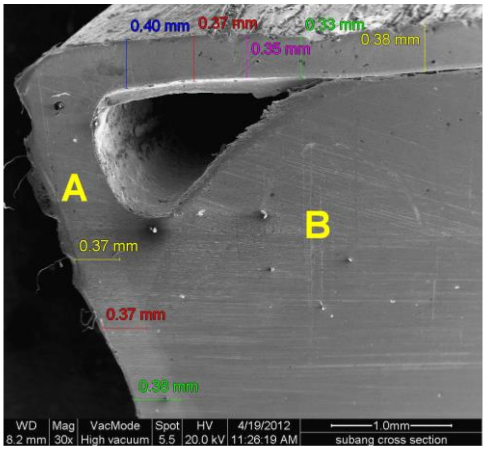
To verify the above-mentioned CT results, the sample was then evaluated by using conventional XRF technique. The XRF test was conducted at INPUT YaPEIM's laboratory using an EDX600 XRF spectrometer. This was done on a few selected positions on the surface of the earrings, and an average value was calculated. Results of the elemental analysis revealed that the outer layer consists of 91.9% Au, 2.019% Ag and 6.081% Cu.

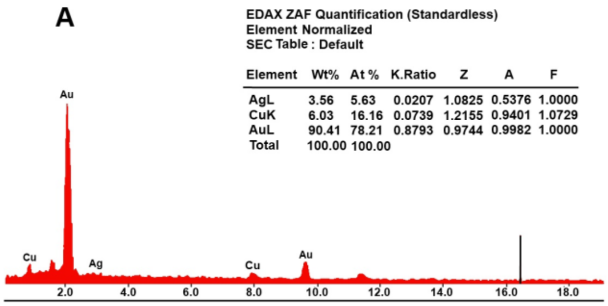
The purity or fineness of gold in a jewellery is indicated by its carat number. As stipulated in the World Gold Council [12] publication on gold jewellery, 24 carat (24K) gold is pure gold. 24K gold is also called fine gold and it is greater than 99.7% pure gold. Proof gold is even finer, with over 99.95% purity, but it is only used for standardization purposes and is not available for jewelry. 22K gold contains 22 parts gold and 2 parts of another metal(s), making it 91.7% Au. 20K gold contains 20 parts gold and 4 parts of another metal(s), making it 83.3% Au. 18K gold contains 18 parts gold and 6 parts of another metal(s), making it 75.0% Au, and so on. Based on this standard composition of gold alloy, the outer layer of the earrings is identified as 22K gold.

The earrings was then sectioned for further analysis. Figure 4 shows a photograph of the earrings after being sectioned into two parts. As can be seen, the inner part is absolutely consists of different metal as compared to the material of the outer layer. It is shiny and the colour is just like pure silver metal. Large air voids are also visible at certain positions between these metals, in particular, at the top and the bottom edges of the earrings. The thickness of the outer layer was measured by a high precision caliper (micrometer) and the average value is about 300µm. The findings are quite consistent with the results obtained by the dual energy X-ray micro CT.

The final test was conducted by using a scanning electron microscope (SEM) equipped with energy-dispersive X-ray spectroscopy (EDX) at the Malaysian Nuclear Agency, Bangi, Selangor. The SEM-EDX used for determination of elemental composition, and the X-ray characteristic spectra observed on the sectioned surfaces are given in Figure 5 (A) and (B) for outer layer and inner part, respectively. In average, the outer layer consists of about 91.5% Au, 2.47%Ag and 6.03%Cu while the inner part contains 100%Ag [13, 14]. These values are quite consistent and in good agreement with the XRF data. The thickness of the outer layer was accurately measured at a few selected locations and the value is between 288-300 µm. All these data confirm the dual energy X-ray micro computed tomography findings.

Figure 4 Photograph of the earrings after being sectioned into two pieces





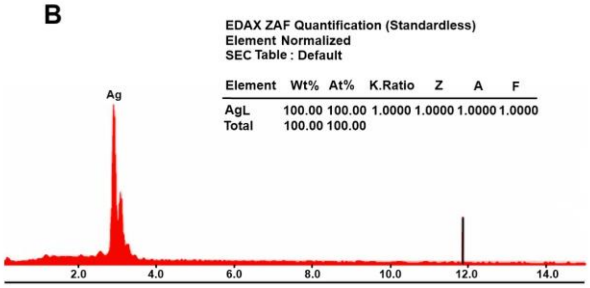


Figure 5 Results of the SEM-EDX analysis of the sectioned surface after it has been polished and cleaned with distillated wáter. Spectrum in A represents elemental composition of the outer layer and spectrum in B corresponds to elemental composition of the inner part

■4.0 CONCLUSIONS

Dual-energy X-ray micro-computed tomography is a promising non-destructive tool for assaying the purity and fineness of gold jewelleries, and thus paving a new way to prevent trading of fake jewelleries. The technique, mathematically combining two tomographs acquired at two distinct energies, allows to obtain both density and atomic number, thus to provide information about material composition, or at least to improve image contrast. Results in this work have shown that the imaging system is capable of producing detailed images of the internal structure of fake gold earrings. The experimental results have been verified by sectioning the sample, then followed by detailed SEM-EDX analysis. The dual-energy approach in combination with an advanced image analysis should provide promising solutions addressing new applications in the future. The limitation of conventional XRF technique for characterising jewelleries has also been demonstrated in this work. To the best of our knowledge, the work presented in this paper is the first of its kind and there is no similar work has been reported in the open literatures.

Acknowledgement

The authors (JA) would like to thank the management and staff of the Malaysian Nuclear Agency, INPUT YaPEIM and Fraunhofer EZRT, Germany for their great help and support. The author (JA) would also like to thank top management of INPUT YaPEIM for providing the financial support and travel grant to carry out this study. This on-going project is part of a long term program to establish the YaPEIM's Non-Destructive Assaying Centre (NDAC) for gold.

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