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Research Paper

An assessment of alloying elements and hardness in gold and silver jewelleries

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Abstract. To exploit the technology of the quality of gold and silver jewelleries, they were subjected to analysis for their alloying composition and their hardness. The compositions were determined by suing X-RF spectrometric method and Rockwell Hardness; Tester on B-scale. The elemental constituents showed that gold jewellery contains Ca (0.73%), Cr (0.13%), Fe (0.33%), Ni (0.09%), Cu (1.90%), Pd (1.70%) and Au (84.90%) while silver jewellery contains Si (1.00%), Ca (0.001), Cr (0.069%), Fe (0.21%), Ni (0.25%), Cu (4.19%, Zn (1.36%), Y (0.5%) and Ag (92.80%) as their oxides. The result of the compositional analysis showed that the jewelleries have heavy elements and when these elements are used as alloying metals in jewelleries they seem to have adverse effects on the users. Other elements might be additions as grain refiners, de oxidizers impurities that help in improving the mechanical properties of the jewelleries. The hardness result showed gold (65 HBR) to be harder than silver (61.7 HBR). Pearson correlation of the metals at 95% confidence with hardness showed that all the jewellery types studied indicated lack of correlation. However, this could not be clearly supported by the plots of the various metal concentrations in the jewelleries against their hardness as the plots of hardness against percent metals suggested both positive and negative correlation. Some metal associated with the jewelleries are soft metals as such they could be prune to scratches and the pores created may habour bacteria. The presence of trace metals or heavy metals which are toxic in the jewelleries may also pose a serious health hazard and potential dangers associated with wearing of jewelleries by unsuspecting users.

Keywords: Alloying elements, jewelleries, hardness, toxic elements.

INTRODUCTION

People have taste for fashion, different metals and non-metals are used in the production of ornaments and jewelleries that require partnership in art and science, thereby utilizing its vast resources in partnership with artisans who demand nothing less than perfection (Jorn, 1966; Encyclopaedia Americana,). Individual metals have different applications at different times and moments. The use of any metal is based on its properties and its alloying elements. Therefore, the various metals used in jewellery could be categorised as precious metals and semi-precious metals. Precious metals include gold, silver and platinum, while semi-precious metals include copper, brass, aluminium and many other semi-precious stones such as topaz, quartz and aquamarine. Costume jewellery on the other hand are made of materials that are non-precious and non-metallic such as seeds, bones, feathers, leathers, calabash, paper, stone, glass and other imitation. The materials used in jewellery are very important as the jewellery itself. The skills in selecting materials for jewellery- making is one of the first elements in jewellery production that must be mastered, which has also been observed that if materials used for jewellery production are bad, it takes a longer period of time and energy to come out with a good work (Caplen, 1983). The use of any material for any jewellery making must be divorced from deception. Where there is insincerity goldsmith can used brass or copper to make jewellery, after coating with gold solution, it could be sold as gold since it looks golden. Apart from copper, gold is the only truly coloured metal used in jewellery, being pale yellow, in the massive state, and red or even purple when finely divided (Ross, 1992). Gold is easily worked and it is very attractive in colour, (bright) ductile, durable and malleable. It can be alloyed with other metals to improve the strength, hardness, etc. It is used in coins making, medals, masks, helmets, shield, weapons, for ceremonial purposes and furniture lining apart from jewellery making. Gold can be used for flat-wares, and hollow-wares. It can also be used as rolled gold, or gold leaf when coated on materials to enhance the surface enrichment. Standard gold is 24 carat and is the purest form of it (Kazanas et al., 1979 and Le'Coinbeitler, 2005). Silver has several properties that made it a natural choice for a jewellery and decorative material. It has unmatched whiteness and luster and excellent workability and is highly resistant to corrosion. The type of silver jewelleries include sterling silver with 92.5% silver, and is often referred to as 925; the other one is white and fine silver which have 99.9% of silver and is stamped 999, (Hilliard, 2001; Etris, 1997). Silver is a precious metal that possesses intrinsic market value in addition to artistic value that is added to it in transforming it into the item of jewellery or decorative art. Jewelleries often contain alloy elements. An alloy is made by melting metals and mixing them. An alloy is a solid solution since the metal elements remain distinct, one suspended in the other. Examples of common alloys include alloys of brass (a mixture of copper and zinc), bronze (a mixture of copper and tin and other metals), and sterling silver (a mixture of silver and copper). Silver alloys readily with gold, copper, and many other metals. This allows a variety of alloys from which to select the best suited to the mode of jewellery or art of craft manufacture. The desire and degree of durability, chemical inertness and the desired appearance of the manufactured item can be obtained by varying the proportion of alloying metals and their properties such as colour, strength, corrosion resistance or working properties. This paper assesses the influence of alloying metals on the hardness and quality of gold and silver jewelleries.

MATERIALS AND METHODS

Samples identification and collection

The study was divided into three, namely; samples identification and collection/laboratory analysis. The samples were identified by the name they are called as Gold (Au), and Silver (Ag), jewelleries. The jewelleries were locally purchased and stored in paper envelopes from different Markets in Bauchi metropolis, Bauchi state, Nigeria.

Sample preparation

Samples were wiped clean with absolute ethanol and dried in an oven at 90°C for 30 min. The samples were allowed to cool before being ground using a crushing machine and the powder stored in corked plastic cups.

Determination of hardness

ASTM E18 Rockwell was adopted for the hardness tester on 'B' scale confidence with 5 mm ball indenter minor load of 10 kg, and hardness value of 101.2 HRB was used as standard block in determining the values of the specimen provided. Before the test, the maturing surface of the indenter, plunger rod and test samples were thoroughly cleaned and the test machine calibrated using the standard block. The samples were then placed on anvil which provided support for the test samples. A minor load of 10 kg was applied to the sample in a controlled manner, without inducing impact arbitration and zero datum position was established and then the reading was taken when the large pointer came to rest.

Metal analysis

Ground samples chemical analysis were analyzed on a Mini X-Ray Fluorescence Spectrometer (XRFS) Model 4 Panalytica, at the National Metallurgical Development Center (NMDC) Laboratory, Jos, Plateau State, Nigeria. The analyses were done in triplicate to ensure good representation.

Statistical analysis

Pearson correlation analysis was performed at 95% confidence on the result of hardness of the jewelleries studied and their constituent metals.

RESULTS AND DISCUSSION

The result of the composition of the jewelleries shows that metals hardly exist in pure **s**tate as most often, during solidification. Other metals form part of the metal of interest through co–precipitation, occlusion, or alloy formation. This implies that the quantities o the metals in jewelleries are not uniform. The distributions of other metals in gold and silver jewelleries are shown in Table 1 and 2. The gold jewellery has 84.16% gold suggesting that it might be 20 karat gold jewellery (Helmenstine, 2006). The total percentages of alloying metals are 14.98% of gold jewellery and 6.884% of silver jewellery as their oxides.

The hardness (HER) of the jewelleries result is presented in Table 2. The result shows that there is a relative increase in the hardness of the samples with

Table 1. Alloying Metals in Gold Jewellery (21 karat).

Metal	Ca	Cr	Fe	Ni	Cu	Pd	Os	Au
Compound	CaO	Cr_20_4	F	NiO	CUO	PdO	Os04	Au
Conc.(%)	0.73	0.13	0.33	0.09	11.90	1.70	0.10	84.90

Table 2. Alloying metals in silver jewellery.

Metal	Si	Ca	Cr	Fe	Ni	Ci	Cu	Y	Ag
Compound	SiO_2	CaO	Cr_20_3	Fe_2O_3	NiO	CiO	CuO	YeO_3	AgO
Conc. (%)	1.00	0.01	0.069	0.21	0.025	4.19	1.36	0.045	92.80

Table 3. Hardness of jewelleries.

Samples	1	2	3	Average HRB
Compound	65.0	66.0	65.0	65.0
Conc (%)	61.0	62.0	62.0	61.0

Key: HRB-Rockwell Hardness 'B' Scale

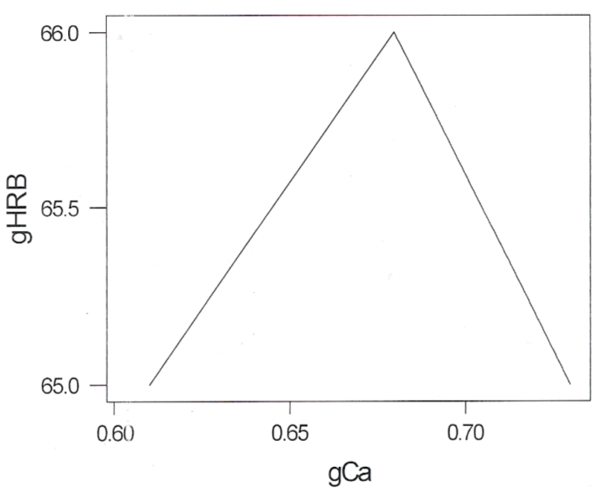

Figure 1. The effect of calcium on the hardness of gold jewellery.

increasing percent concentration of alloying metals (Table 3). This implies that alloying elements have effect on the hardness of the jewelleries. The result of additional alloying elements whose concentration does not exceed 1% could be considered as an additive or an impurity (Ort, 1997). This suggests that Cu (11.9%) and Pd (1.7%) serve as alloying elements in gold jewellery while Cu (4.19%) and Zn (1.36%) provide the alloying elements in silver jewellery. Other elements (Tables 1 and 2) could be considered therefore as additive or impurities.

The correlation of the metals found in the jewelleries using spearman correlation at 95% confidence with all the jewellery types studied indicated lack of correlation. However, this could not be clearly supported by the plots of the various metal concentrations in the jewelleries against their hardness (Figures 1 and 2). Figure 1 provides the effect of Calcium on the hardness of gold jewellery. The plot indicates increasing hardness as the percentage of calcium increases but decreases as it exceed 0.6%; implying that calcium might be needed at levels below 0.675%. Ort (1997) showed that addition of an element may improve one property and have adverse effects on another so that it acts both as a useful addition and an unwanted impurity. Some additives serve as gain refiners and deoxidizers and this has been reported to be unnecessary if alloys melted and cast using proper condition (Ort, 1997). Zinc, silicon, boron and phosphorus act as deoxidizers (Kenneberg and Williams, 1997).

Some metals could be base metals, coating elements, while some may be additives (Rollanson, 1973). The effect of chromium on the hardness of gold jewellery shows that hardness is reduced with the addition of chromium to gold jewellery (Figure 2) until reaches concentration where it remains constant, chromium is increased. The same trend is shown by iron, nickel and copper addition to gold jewellery (Figures 3, 4 and 5). Iron has been reported to act as impurity in gold jewellery (Kenneberg and Williams, 1997). The effect of concentration of palladium on the hardness of gold jewellery follows similar pattern as that of calcium (Figure 6). However, the plot suggests addition of small concentration of palladium may help in sharp increase in hardness of the jewellery. Osmium concentration has a 0.10% limit for the jewellery to remain permanently hard beyond which the hardness will reduce (Figure 7). Increased silicon concentration makes impact on the hardness of silver jewellery only when the silicon levels reaches between 1.00 to 1.10% (Figure 8). The effect of Ca and Cr on the hardness of silver jewellery only becomes

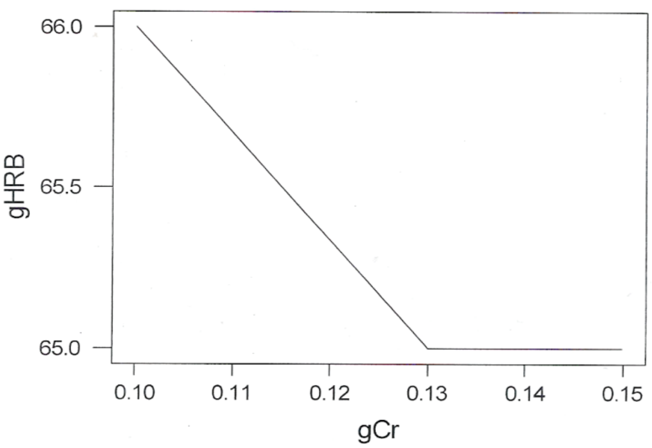

Figure 2. The chromium concentration on the hardness of gold jewellery.

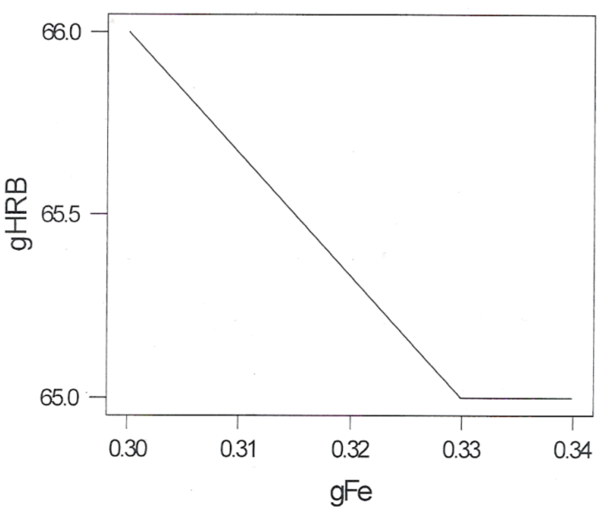

Figure 3. Iron concentration effect on the hardness of gold jewellery.

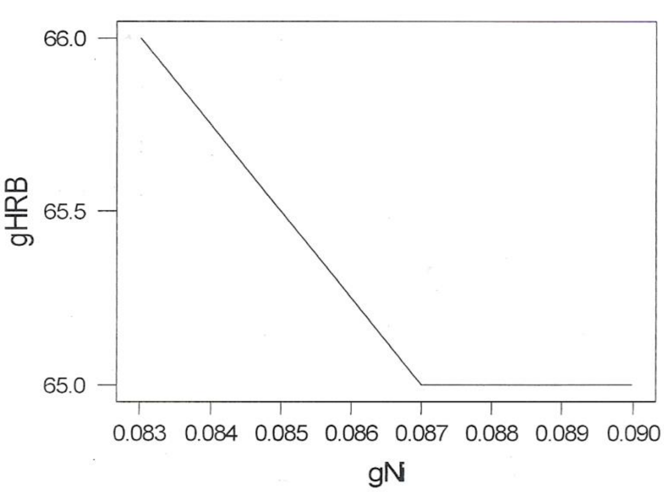

Figure 4. Nickel concentration effect on the hardness of gold jewellery.

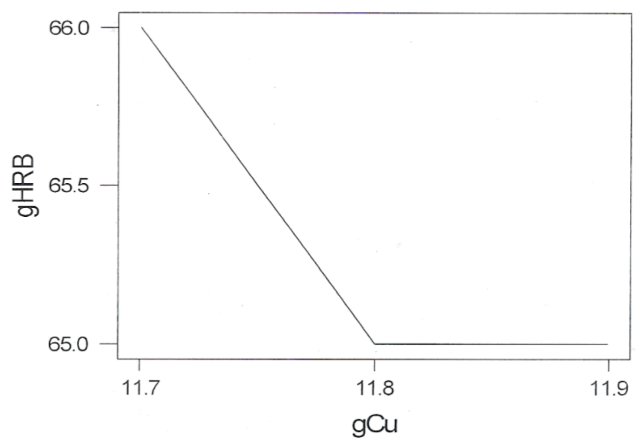

Figure 5. Copper concentration effect on the hardness of gold jewellery.

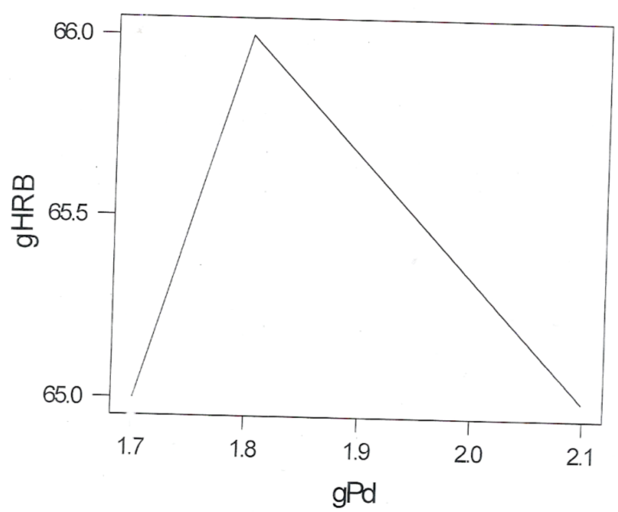

Figure 6. Palladium concentration effect on the hardness of gold jewellery.

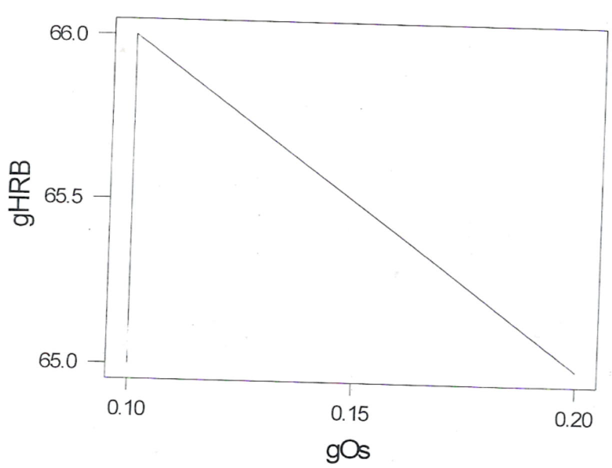

Figure 7. The effect of osmium concentration on the hardness of gold jewellery.

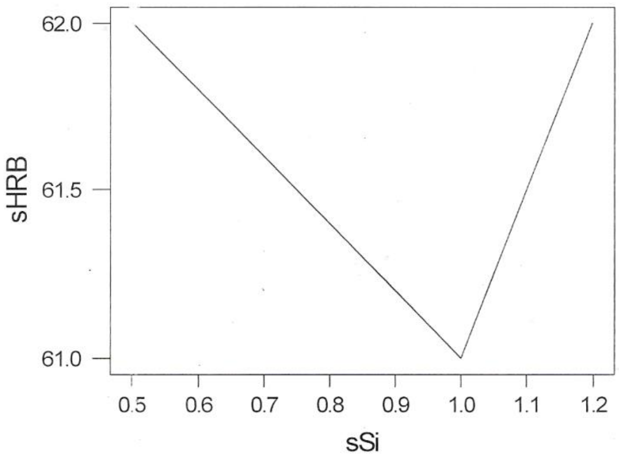

Figure 8. The effect of silicon concentration on the hardness of silver jewellery.

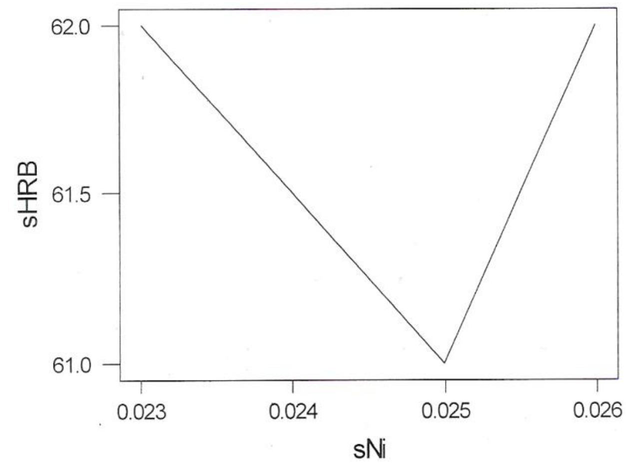

Figure 11. The effect of nickel concentration on the hardness of silver jewellery.

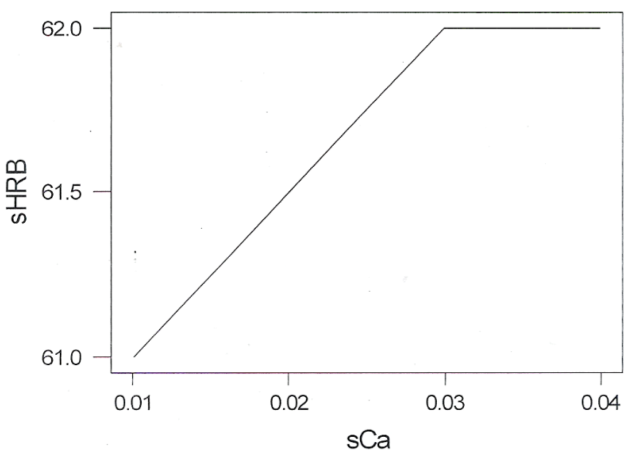

Figure 9. The effect of calcium concentration on the hardness of silver jewellery.

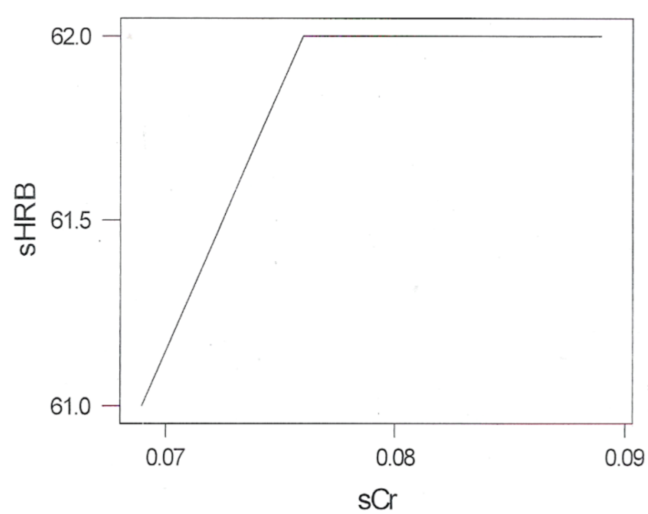

Figure 10. The effect of chromium concentration on the hardness of silver jewellery.

relevant at concentrations less than 0.03 and 0.057%, respectively. The steady rise with increasing concentration of calcium and chromium in silver jewellery becomes steady without increase in hardness at any level beyond 0.03 and 0.075% (Figures 9 and 10). Nickel and copper have similar pattern with that of silicon. Their lower concentrations do not have influence on hardness. At higher levels they cause increase in hardness of silver jewelleries (Figures 8 and 11).

Silver jewellery does not rust or perish, it lasts for a long time and it only tarnishes when it comes in contact with sulphur (perspiration from the body) and other oils and creams (Ogbe, 2000).

The presence of alloying metals in jewelleries serve as builders, refiners, deoxidizers and base metals but much of them may have adverse effect. Heavy metals were found to be present in jewelleries that could cause allergic reaction to the wearers. The implication is that the metals pose a potential health hazard to the society and the environment. Chromium, copper, zinc, nickel and iron that are found in the jewelleries have been identified as toxic to plants, animals and humans (Lenntech, 2006). Zinc (Zn) does not occur naturally in elemental form. It is one of the most mobile heavy metals in surface and ground waters because it has been reported to be soluble at neutral and acidic at (pH) level (Evanko and Dzornbak, 1997). This suggests the tendencies and ease by which Zinc can leave the surface of jewelleries into the environment. Cu mobility is reduced by sorption to surfaces over a wide range of pH (Dzornbak and Morel, 1990). When ingested, it may cause ulceration of gastrointestinal Musocosa, hemolysis, hepatic necrosis, and renal damage (Van Campen, 1991). The release from jewelleries may be through sweet and bathing with the jewelleries worn. The concentration of Cu by this mode may be small but with large numbers of people wearing jewelleries worn the concentration dissolved might be significant ((Lenntech, 2006). Chromium (Cr) mainly is found as metallic Cr or in trivalent 3+ and hexavalent 6+ states. Chromium exposures may occur through the use of chromed jewelleries. Trivalent Cr+ is poorly absorbed through the skin, but hexavalent Cr6+ is readily absorbed and acts as an irritant, causing burns, ulceration and it is the most mobile element in waste disposals (Smith et al., 1995). Nickel represents a potential hazard. No occupational exposures arise from handling metallic objects such as Ni jewellery and Ni coins. A high rate of Ni – allergy is associated with ear piercing and subsequently wearing of Ni alloy jewellery (Berova et al, 1987). Nickel also cause dermatitis, respiratory disorders and inhibits the enzyme cytochrome oxidase and dehydrogenize (Sunderland and Donelly, 1995). Cross–reactivity is an important issue in Ni – allergy as Ni exposure often occurs in the context of multiple metal exposures. The present of Ni as constituent of jewelleries expose man to the risk of ingesting.

CONCLUSION

The jewelleries studied were found to contain one base metal and alloying or coating metals. Some are additive and deoxidizers. In gold jewellery the base metal was 85.9%, while Cu was the coating metal with 11.9%. The percent of gold suggests that it might be 20 karat gold jewellery (1 karat = 4.16% gold) and this level of gold in jewellery has been found to be appropriate for body jewellery (Helmenstine, 2006). Similarly, Silver jewelleries have 92.8% as base and Cu (4.19%) as the coating element. The result of the compositional analysis showed that the jewelleries have trace elements and heavy metals. When these trace/heavy metals are used as alloying metals in jewelleries they seem to have adverse effects on the users (Mc Cluggage, 1991; INECAR, 2000; Fermer, 2001 and Lennetech, 2004). Other element might be additions that help in improving the mechanical properties if the jewelleries or could be there as impurities. Some people are sensitive to the metals present in Karat gold such as Ni, Ag, Zn and Cu. Similarly, while gold has been thoroughly investigated to often cause more adverse reactions than yellow gold because of the high amount of Nickel which causes the white colour. The relatively high copper level in the gold jewellery may be a cause for concern as perspiration could be acidic enough to corrode the copper which is often evident by the blackening of the jewellery and discoloration of the surrounding skin (Helmenstine, 2006). Some of the components of the jewellery were found to have hardening properties. These properties help to reduce abrasion, tearing and scratching. Silver jewellery is made of predominantly silver, with copper constituting the next major element. Silver is very soft and is easily scratched, this may reduce the quality of the jewellery and the pores may trap bacteria which will encourage infection (Helmenstine, 2006). It is advised that silver jewellery can be safely worn in healed pierced ears and should not be worn on unhealed pieced or near a moist area of the body such as the mouths or the genitals. Silver does not appear to harm the body except by castings blackening of the tissue (Lewton–Brain, 1999). Nickel was found in all the jewellery types and it has been associated to dermatitis allergy. These jewelleries all contain chromium which is also a toxic metal could serve as a source of chromium ingestion. Some metals associated with the jewelleries are soft metals as such they could be prune to scratches and the pores created could habour pathogenic bacteria. The presence of these trace metals or heavy metals which are toxic in the jewelleries may also pose a serious hazard to the wearers. The potential danger associated with the qualities of these alloying elements is that the wearers may be a potential victim for skin rashes and irritations if it contains nickel when worn by unsuspected users.

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