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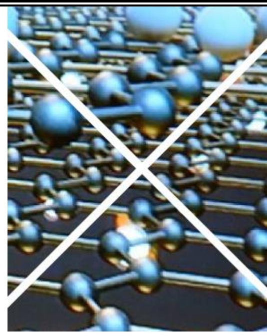

INFLUENCE OF TERNARY ALUMINIUM AND QUATERNARY ZIRCONIUM ON THE PHYSICAL PROPERTIES OF BELL METAL

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Abstract

The change in physical properties of bell metal due to ternary addition of Al and quaternary addition of Zr has been reported. Cast samples are aged both isochronally and isothermally. Investigation on the age hardening property reveals that due to the formation of intermetallic precipitates, significant hardening takes place in case of Al added alloys and Zr addition enhances the thermal stability of the alloy. The comparative acoustic response study discloses that heat-treatment improves the sound quality of all three bell metal alloys. The base alloy shows highest decibel values for differently aged temperatures. The ternary Al added alloy and quaternary Zr added alloy show continuous increase in decibel values with increasing ageing temperature. However, the acoustic response curve of the alloys almost followed the similar pattern of hardness curve with the ageing temperature. Spectral reflectance study reveals that the ternary Al added alloy shows better percent reflectance with the increase of wavelength of incident light. The quaternary Zr added alloy is also seen to retain its superior optical properties for all ageing conditions. It is found from microstructural analysis that ternary Al addition creates a new microstructure with relatively large dendritic arms and the quaternary addition of Zr refines the grain structure. The base alloy attained almost full recrystallization state after ageing at 300°C for 60 minutes while the ternary Al added alloy was also recrystallized at elevated temperature of 500°C but the quaternary Zr added alloy did not attain recrystallization and maintained its grain structure due to thermal stability.

Keyword: Bell metal, Age hardening, Sound level, Reflectance, Microstructure.

1. Introduction

Since the dawn of civilization copper and copper-based alloys have been utilized in quite a variety of applications. Copper can form alloys more frequently than most other metals and with a wide range of alloying elements to produce the alloys like brass, bronze, bell metal, beryllium copper, copper nickels etc. providing excellent high-temperature strength [1-3]. Bell metal is an alloy of copper and tin which is more rigid and tough than either of the alloy material constituents. Historically it has been used for making bells and related cymbal type percussion instruments. That is why the copper-tin alloy which has a higher tin content than regular bronze is called bell metal. It has acute resistance property against the atmospheric corrosion. The antique blacksmiths could easily cast this metal due to its low melting point and so various shapes could be formed with less effort. Bell metal possesses attractive golden color however some other metal constituents can refine its color, finish or improve the flow of liquid melted alloy better [4, 5]. Bell metal is famous for its characteristic of high resonance during impact. It has been used since prehistoric period to create sound of high resonance. The higher composition of tin increases the rigidity of the metal which increases the resonance [6, 7]. Now bell metal is used in different musical instruments along with different religious offerings and rituals, making forge tools, weapons, cannons, vessels, utensils, statues and ornaments etc. Industrially it is used for bearings, bushings, piston rings and valve bodies [8, 9]. Bell metal is actually a form of bronze with a higher content of tin, sometimes usually in approximately a 4:1 ratio of copper to tin. The bronze alloy with higher tin constituent is commonly utilized in variety of industrial application like fabricating and manufacturing bushings, bearings, gears, ship and pump impellers and so on where they are subjected to high friction, heavy load and sliding application. The applications also include building portable bridge components and turntables for bridges. Although tin offers a negative factor on the properties of electrical and thermal conductivity of the alloy, it is comparatively more influential than zinc specially in strengthening applications. Age hardening is a methodology of copper strengthening where the system offers to convey strength to the materials and their alloys [10]. The material is allowed to be heat treated at elevated temperature so that the solutes can be dissolved and then quenching the metastable supersaturated solid solution. Thereafter reheating to intermediate temperature allows the profuse solute to induce precipitate which substantially forms second phase constituents. The addition of trace zirconium with copper, results in maintaining excellent electrical and thermal conductivity [11]. If aluminum is added to copper based alloys, various intermetallic precipitate such as Al_2Cu forms which increases the hardness [12]. Moreover, aluminum has excellent corrosion resistance, convenient malleability, high ductility, non-magnetic, non-toxic, light weight and lower cost [13]. On the other hand zirconium has grain refining ability [14]. In view of the importance of trace additions of zirconium in the high tin bronze, the age hardening studies can also be conducted with quaternary addition. This addition of zirconium increases thermal stability of the material by refining the grains.

This study is about improving the physical properties of bell metal by adding ternary aluminum and quaternary zirconium. From the literatures there is hardly any study available on the effect of aluminum and zirconium on the physical properties of bell metal. In this paper the effects of these additions along with the analysis of isochronal and isothermal age hardening, acoustic and optical characteristics and microstructural properties are observed on the evolution of microstructures in experimental alloys.

2. Materials and Methods

Three samples of bell metal, ternary aluminum added bell metal and quaternary zirconium added bell metal were fabricated individually through continuous casting and melting process. Copper, tin and aluminum in the pure form were used for the alloy preparation where zirconium was taken in the form of and aluminum-zirconium master alloy. The casting was done in a crucible of clay-graphite. The furnace used was conventional fire pit type using natural gas as fuel. To fabricate bell metal Alloy 1, copper and tin were melted in the clay-graphite crucible. For making Alloy 2 aluminum was added by dipping it into another molten metal of copper-tin. Consecutively Al+10wt% Zr master alloy was added to the molten copper-tin for making the Alloy 3. All three alloys were casted individually. The melting temperature was tried to be keet at 1300°C more or less. Three molds of stainless steel with a size of 20× 150 × 150 mm^3 were preheated before pouring the cast into it. A mixture of water-clay was made and them as inner layer of the preheated molds. The individual melts were then poured into those preheated molds. The chemical compositions of the alloys were characterized by Optical Emission Spectroscopy method. Table 1 shows the chemical compositions of all three alloys.

Through machining process the cast samples were burnished so that the oxide film could be removed from the exterior and then different samples were prepared from the castings where the sample size was 15 x 15 x 4 mm^3. Then the fabricated alloy samples were heat treated for different time durations at variety of temperatures. The emery papers of various grits were used to polish the surfaces of the heat treated samples. A hardness tester machine named Micro Vickers Hardness Tester was used to find the micro-hardness of the heat treated samples. Several indentations were taken from different spots of each sample surface where indenter was allowed to apply 1Kg force for the duration of 10 seconds.

Three metal plates were also obtained with the size of 90 x 60 x 10 mm^3 from individually cast bell metal alloys through machining process. They were also aged at different temperatures for one hour. A wooden platform was made for the experiment to investigate the sound intensity level of all these bell metal plates. The differently aged metal plates were hanged through a pair of chain and a bearing ball of stainless steel was used to strike on the metal plates to create sound. The set up was made in such a way that the ball would strike the plate with equal force each time. An industrially usable digital sound level meter of model "Digital Sound Level Meter AS804" was used to measure the sound intensity level. To get accurate result a Digital Sound Calibrator was used to calibrate the digital sound level meter. At least 100 decibel values of each of the alloys were taken to get more precise values of sound intensity.

The reflectance test was carried out with the UV-VIS-NIR Spectrometer. The aged samples were machined to smooth powder to perform this test. The optical microstructures of the alloy samples were characterized in the conventional way. A wet polishing machine with velvet clothed wheel with the addition of alumina powder was used to make scratch free polished surface. Acetone was applied to clean the surface. Mixture of Hydrogen peroxide (3%) and typically suggested one of Ammonium Hydroxide were taken in the ratio of 1:1 as a metallographic etchant. The samples were then washed and dehydrated. For microstructural analysis several photomicrographs were captured with a variation in magnifications through OPTIKA microscope. A scanning electron microscope JOEL along with an energy dispersive X-ray analyzer (Model: Link AN - 10000) were used for mapping the individual phases in microstructures. The samples used to find the SEM and EDX were heat treated to 300°C and etched with same etchant solution used in finding the microstructures.

Table 1. Chemical Composition of the Experimental alloys (wt%).

	Sn	Pb	Fe	Ni	Al	Si	Cr	Zr	Mn	Cu
Alloy 1	24.935	0.000	0.000	0.019	0.005	0.001	0.004	0.000	0.001	Bal
Alloy 2	25.444	0.000	0.012	0.018	1.165	0.007	0.003	0.000	0.001	Bal
Alloy 3	25.008	0.010	0.020	0.018	1.170	0.002	0.004	0.240	0.002	Bal

3. Results and Discussion

3. 1. Isochronal Ageing

The results of isochronal ageing of the cast bell metal Alloy 1, ternary Al added Alloy 2 and quaternary Zr added Alloy 3 at different temperatures for 60 minutes are shown in Fig. 1. From the figure, it is seen that Alloy 2 and 3 have shown substantial ageing reaction. However, Alloy 1 is seen to have a continual alleviation of hardness with increasing ageing temperatures but small variation is observed beyond 300°C. It is known that copper when alloyed with tin, forms tin bronze and the hardness increases with the increase in tin content of the alloy. This softening is the effect of stress relieving, grain coarsening and recrystallization of the alloy respectively. But, when aged at higher temperatures the hardness of the bell metal increased moderately due to grain growth [15, 16].

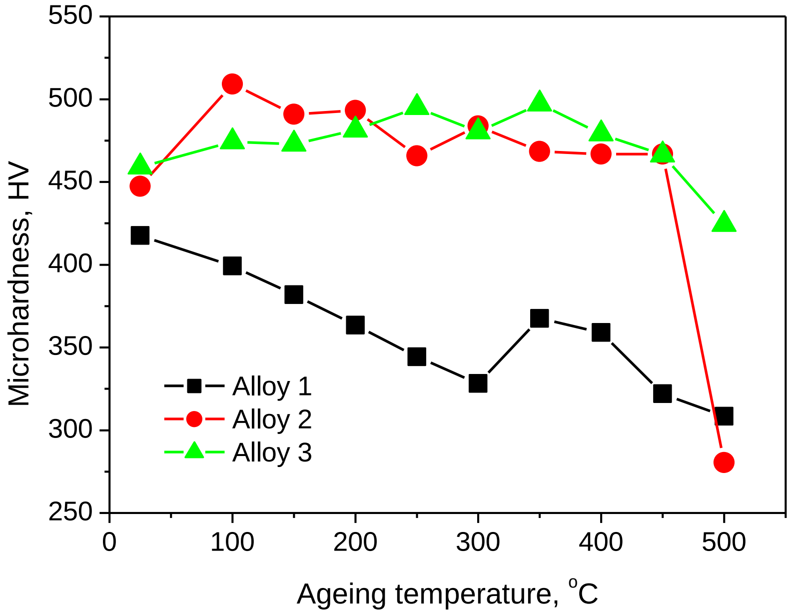

Figure 1. Isochronal ageing curve of the alloys, aged for 1 hour

Alloy 2 and Alloy 3 have shown better response to the ageing effect and retained their hardness up to 450°C. The results of the experiment suggest that the precipitation hardening shown by the alloys was because of the addition of aluminum. It is well established that addition of small amount of Al to Cu alloy increases the hardness of the alloy [17]. During solidification and ageing, various intermetallic phases are formed by reaction between Al and Cu. The major intermetallic precipitates which influence the hardness of the alloys are Al_2Cu, Al_4Cu_9 and Cu_3Al_2 [18, 19]. Beyond 450°C there was a steep drop in hardness of Alloy 2. The reason behind this phenomenon is the precipitation coarsening and recrystallization effect on the alloy. It appears on close observation that Alloy 3 with quaternary addition of Zr showed the highest resistance to age softening. The quaternary addition of Zr has increased the thermal stability of the alloy. The primary reason behind this is the formation of intermetallic compounds such as Al_3Zr. Dispersoid particles of Al_3Zr are formed during ageing of Alloy 3. These particles hinder recrystallization and grain growth by restraining grain boundaries during successive heat treatments of the experimental alloy [20]. This hindrance in grain coarsening of the alloy increased its thermal stability. Thus, Alloy 3 maintains high hardness in over-aged situation.

3.2. Isothermal Ageing

The hardness differential of the cast alloys when they were isothermally aged at 300°C for different time periods are shown in Fig. 2. Similar trend as isochronal ageing is seen in the alloys after being heat treated isothermally for up to 8 hours. From the graph it is seen that Alloy 2 and 3 has shown high hardness in this temperature. The reason for this is the formation of the intermetallic compound of copper aluminate as discussed earlier. During the ageing process of the alloys, the formation of intermetallic precipitates significantly strengthened the alloys which led to the peak hardness [21, 22]. It is seen from the graph that the base Alloy 1 did not react positively to the isothermal ageing treatment. The alloy hardness dropped with the ageing period and a sharp decline in hardness was found after 30 minutes of ageing. There was again a steady decline in hardness after 4 hours of ageing process. In contrast, Alloy 2 and 3 started hardening from an early time. Both the alloys reached their hardening peak after 60 minutes of ageing. After attaining their peak hardness, Alloy 2 and 3 maintained a more or less a constant value of hardness. The maximum hardening due to alloying additions was found after 60 minutes of ageing.

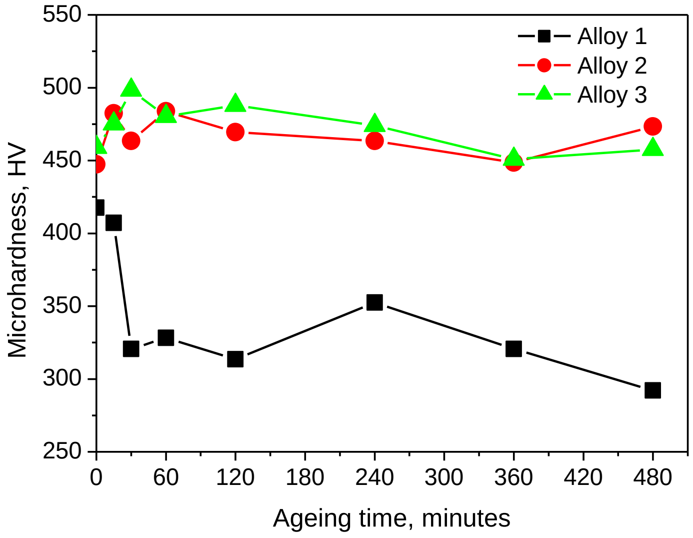

Figure 2. Isothermal ageing curve of the alloys, aged at 300°C

3.3. Acoustic Response Properties

Fig. 3 indicates a comparison of sound intensity level among all three experimental alloys. Clearly the base Alloy 1 creates highest sound intensity among them. This is because the unique chemical bond which holds the copper and tin together, has the resisting ability of forming cracks which creates mild vibration when struck with the striker [23]. Atoms being arranged in highly ordered structure form crystal. When copper-tin crystal is struck hard, copper can cave in because of its softness. That's why tin atoms are needed to replenish the fragile spots of its crystal. As a result, it will resonate the sound due to the strike as opposed to being collapsed. Moreover the crystal lattice formation of this alloy has high impact absorption ability [24]. This results in a resonant pitch that causes the metal to vibrate strongly and smoothly in a complex nodal system. The other two alloys showed lower decibel values because the ternary aluminum addition and the quaternary zirconium addition create different metallurgical composition which affects the optimum balance of the mutual weight percentage of copper and tin. The increasing weight percentage of other materials except tin creates this sound decay [7, 25]. The ternary addition of aluminum and quaternary addition of zirconium also change the microstructure which reduce the self-sustained oscillating ability of the base alloy. Different intermetallic precipitates of Al_4Cu_9, Cu_3Al_2 and Al_2Cu form in aluminum added alloys and disrupt the copper-tin bond which was previously responsible for the higher resonance whereas in case of zirconium added alloy Cu_9Zr_2 and Al_3Zr intermetallics are accountable for interrupting such attractive sound.

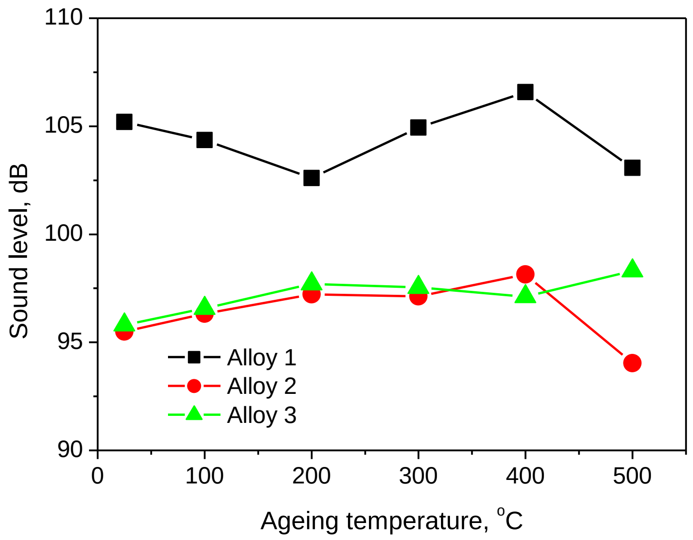

Figure 3. Variation of sound level of the alloys as a function of ageing temperature

Fig. 3 also shows individual acoustic response at different ageing temperature. The sound intensity curve of Alloy 1 shows that the cast bell metal can withstand its high pitch even at elevated temperatures. It happens due to the unique abilty of cast bell metal to maintain its sound pitch level with ordinary changes in temperature [26]. It is also seen that the sound intensity curve of Alloy 1 followed almost similar trend of the hardness curve. Therefore it can be rationally expected that due to heat treatment at differently ageing conditions, the change in hardness related to the bell metal alloy can be considered as a relevant factor influencing sound intensity of the alloy [27]. From Fig. 3 it is seen that bell metal showed slightly lower decibel value while it tends to have precipitation coarsening and dissolution of some phases at the temperature of 200°C but again reached at higher decibel value at 400°C. The formation of new set of defect free grain after recrystallization is the reason behind this higher decibel value at elevated temperature. The other two alloys individually also showed better acoustic performance at elevated temperature compared to their initial situation. Ternary aluminum added bell metal Alloy 2 showed continuous increase from its initial value in acoustic response during impact with the increase in ageing temperature up to 400°C and after that it also suddenly dropped which follows the pattern of hardness curve discussed in the previous investigation. This occurred because of precipitation coarsening and recrystallization effect. Alloy 3 showed better response than Alloy 2 in such a way that it not only shows continuous increase in acoustic response with respect to ageing temperature but also tended to withstand at elevated temperature of 500°C. The quaternary addition of zirconium is the reason behind this better sound response. Zirconium provides resistance to softening with ageing temperature and allows thermal stability. All three alloys showed better sound intensity value compared to their initial state. Age hardening is responsible for improving the quality of acoustic response of all three bell metal alloys [28].

3.4. Reflectance Behavior

Spectral reflectance curve is a plot of reflectance of the cast alloys as an outcome of wavelength. Fig. 4 displays the change of reflectance of the experimental alloys, after being aged at 300°C, with the wavelength ranging from UV to the Infrared region. From the graph it is seen that the reflectance of the alloys increase gradually with the increase of the wavelength. At any given wavelength, Alloy 3 showed the highest value of reflectance whereas the base alloy showed the lowest. As the wavelength increases the refractive index becomes smaller which leads to increase in reflectance [29]. It is clear from the reflectance spectrum curves that the addition of aluminum to the base alloy resulted in the increase of reflectance. This is because at a given wavelength, aluminum increases the reflectivity of copper alloys when it is added [30, 31]. From the figure it is seen that the addition of zirconium has increased the overall reflectance of the alloy. Zirconium typically shows excellent optical properties and has higher reflectivity when added as an alloying component [32].

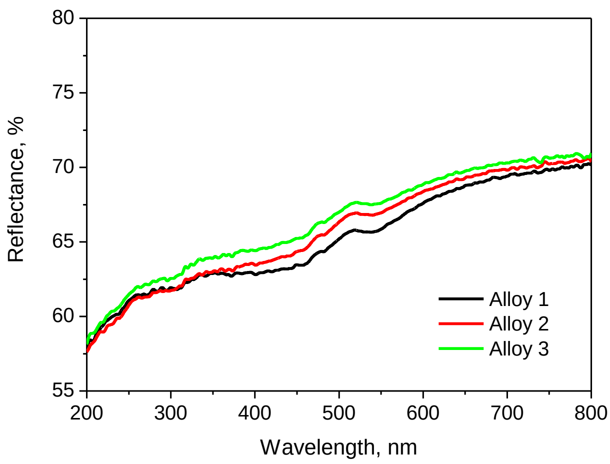

Figure 4. Reflectance behaviour of the cast alloys as a function of wavelength

The variation of mean spectral reflectance with respect to the ageing temperature of the experimental alloys is displayed in Fig. 5. It is seen from the figure that the reflectance of the experimental Alloys 2 and 3 are considerably higher than that of Alloy 1. It is due to ternary addition of aluminum and quaternary addition of zirconium respectively [31, 32]. It is also seen from the figure that the variation of reflectance due to ageing temperature are almost similar for all three alloys up to 400° C. But at 500° C there is a notable change in the variation trend of the experimental alloys. The reflectance of Alloy 3 increased while the reflectance of Alloy 1 and 2 decreased to some extent. It is due to the change in grain structure of Alloy 1 and 2 at 500° C. There are notable changes in the reflectance of material due to change in its grain size [33]. The overall reflectance of material decreased due to increase in its grain size [34]. It is also seen that grain coarsening as well as grain growth occurred in Alloy 1 and 2 at ageing temperature of 500°C (Fig. 7). This augmentation in grain size has resulted in the alloys showing decreased reflectance value at that temperature. The addition of zirconium to copper-based alloys causes grain refinement of the alloy thus decreasing the grain size of Alloy 3 [35]. This also contributes to the increased reflectance value seen in Zr added Alloy 3 at all ageing temperatures.

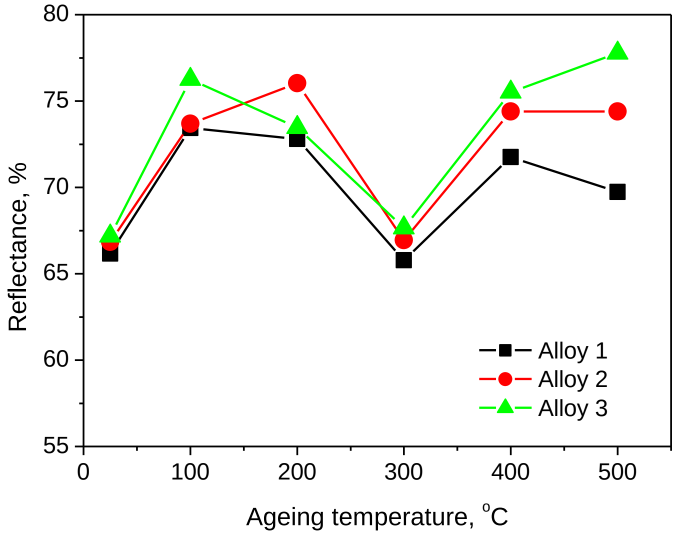

Figure 5. Variation of reflectance of the cast alloys with ageing temperature at 520nm wavelength

3.5. Optical Micrographs

The microstructural features of an alloy significantly influence its mechanical properties. Fig. 6 displays the optical micrographs of the experimental alloys. It is seen that there are visible changes in microstructures because of difference in chemical composition of the alloys. The micrographs showed relatively coarse non-uniform grain structure. The microstructure of Alloy 1 is seen to be comprised of islands of α phase particles with needle like β phase along the grain boundaries (Fig. 6a) The tin content of the high-tin bronze of Alloy 1 is slightly less than the required amount to make a complete β phase [36]. A complete change was observed in the microstructure of Alloy 2 due to the addition of Al (Fig. 6b). At room temperature it showed long dendritic arms which is different from the island-like grain structure of Alloy 1. The long dendritic arms in the microstructure of Alloy 2 is directly caused by the presence of Al in the alloy [37, 38]. There were also visible difference in the microstructure of Alloy 3 (Fig. 6c). The shape and structure of the dendritic arms seemed to be much more refined. The reason behind this is the grain refining effect caused by the addition of Zr to the alloy. Zirconium is a grain refiner and is commonly known to cause grain refinement of copper alloys when added in small amounts. The zirconium present in Alloy 3 reacted with the copper to form an intermetallic Cu_9Zr_2 [39]. Another intermetallic compound formed by zirconium with the aluminium present in the alloy is Al_3Zr. The grain refinement was caused by the presence of Al_3Zr particles that act as effective heterogeneous nucleation point preventing the grain growth during solidification of the Alloy [40]. Grain growths were also restricted due to the generation of Cu_9Zr_2 precipitates [41]. The existence of these intermetallic precipitates refined the microstructure of the alloy and increased its thermal stability [42].

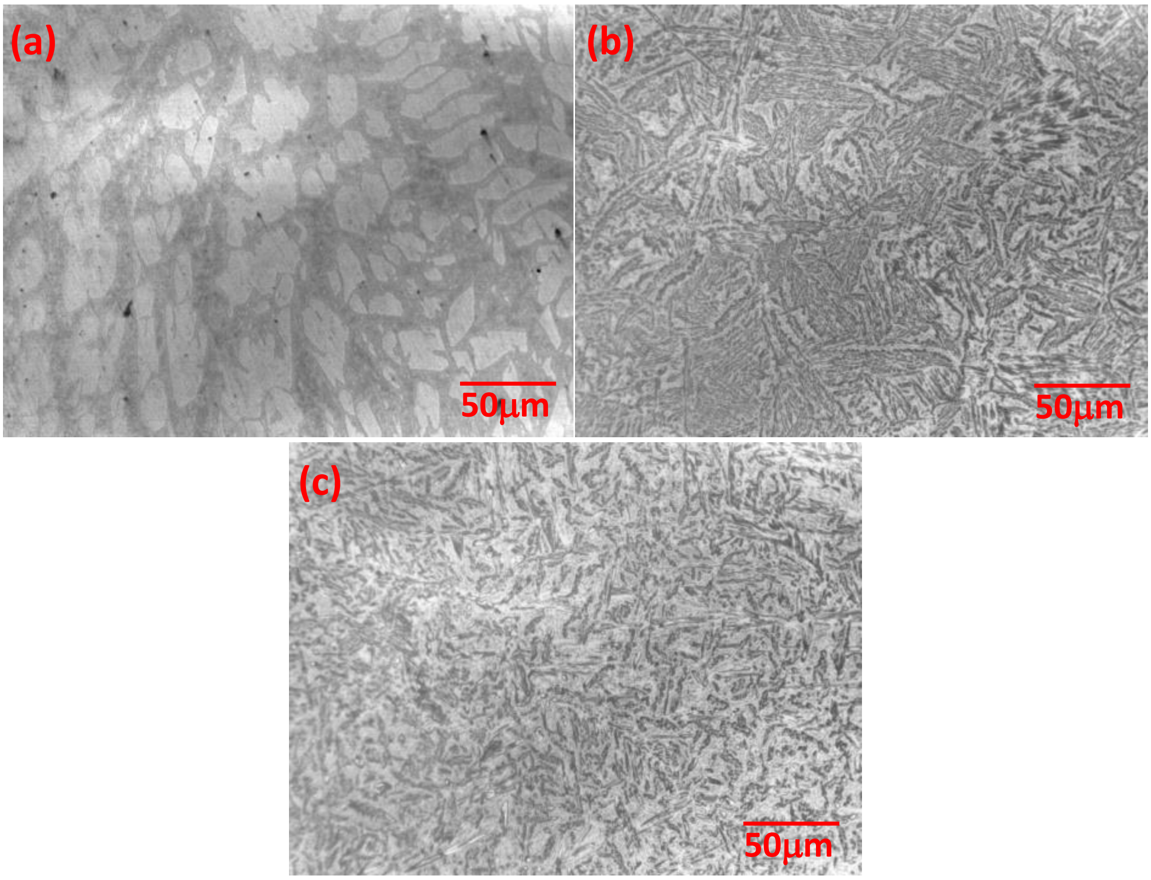

Figure 6. Optical micrograph of the experimental as cast alloys a) Alloy 1, b) Alloy 2 and c) Alloy 3

Fig. 7 displays the optical micrographs of the cast alloys after they were aged to 500°C. The micrographs show that there were substantial change in the microstructure of Alloy 1 and 2. The grains of Alloy 1 was seen to be fully recrystallized (Fig.7a). The recrystallization of the grains is the main reason behind the reduction of hardness during the ageing process of the alloy. The micrograph of Alloy 2 also showed that there has been complete recrystallization of the dendritic arm structure (Fig.7b). This corresponds to the sudden softening of the alloy when aged at 500°C. The recrystallized grains of Alloy 1 and 2 were much more visible under the metallurgical microscope and the original grains were replaced by a new set of defect-free grains. Ageing at 500°C for 1 hour did not cause any significant change to the grain structure of Alloy 3 (Fig. 7c). The dendritic arm structure remained refined as before, although there was partial recrystallization. This adherence of the refined grain structure of Alloy 3 directly corresponds to the thermal stability of the alloy provided by the formation of metastable Cu_9Zr_2 and Al_3Zr [41, 43].

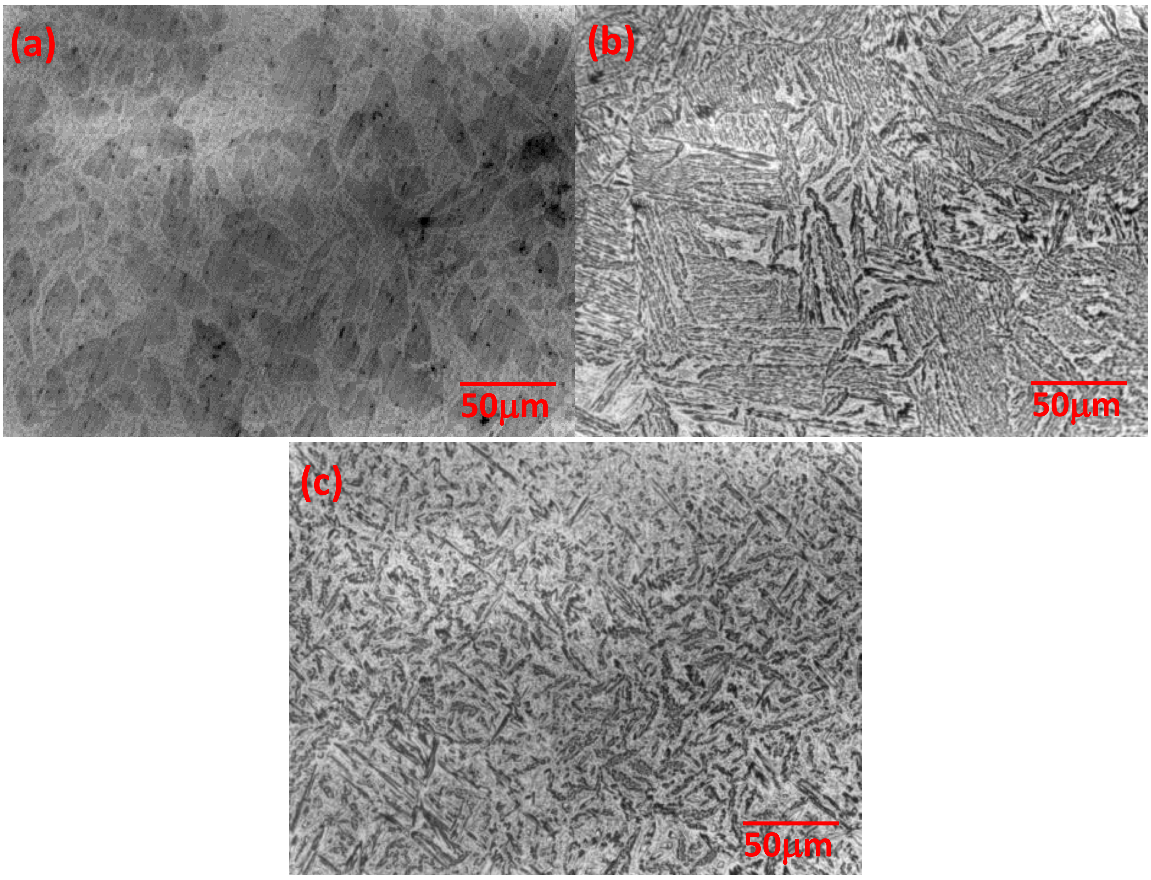

Figure 7. Optical micrograph of the experimental alloys aged at 500°C for one hour a) Alloy 1, b) Alloy 2 and c) Alloy 3

3.6. SEM Micrographs

The SEM micrographs of the experimental alloys after being annealed at 300 °C at 200X magnification level are represented in Fig. 8. From the SEM micrograph of Alloy 1 it appears that the grains of the alloy appear to be recrystallized (Fig. 8a). This observation is in correspondence to the drastic drop in hardness of the alloy when it was aged at 300°C for 60 minutes. The weight percentage of elements obtained from EDX analysis of the SEM is 69.83% Cu and 30.17% Sn. From the SEM micrograph of Alloy 2 it is seen that there are coarse dendrites with considerable amount of second phase constituents present in the inter-dendritic phase (Fig. 8b). The appearance of the coarse dendrites is due to the presence of aluminum in Alloy 2 [18, 19]. The weight percentages of elements obtained from corresponding EDX analysis of the SEM are 1.03% Al, 59.41% Cu and 39.55% Sn. The grain structure of Alloy 2 did not appear to be recrystallized. This directly corresponds to the high hardness retention of the alloy at 300°C. The SEM micrograph of Zr added Alloy 3 showed that there occurred a substantial refinement of the dendrites and the second phase constituents have been decreased in quantity (Fig. 8c). The inclusion of Zirconium to the alloy is seen to have appreciable grain refinement effect on the dendrites that appeared in the microstructure of Alloy 2. During the solidification of Alloy 3, growth restrictions occurred in the nucleant particles introduced by the addition of Zr to the alloy. These growth restrictions are the main reason for the grain refinement of the alloy [20, 44]. In case of Alloy 3 the weight percentage of elements are 1.18% Al, 61.81% Cu, 36.43% Sn and 0.58% Zr.

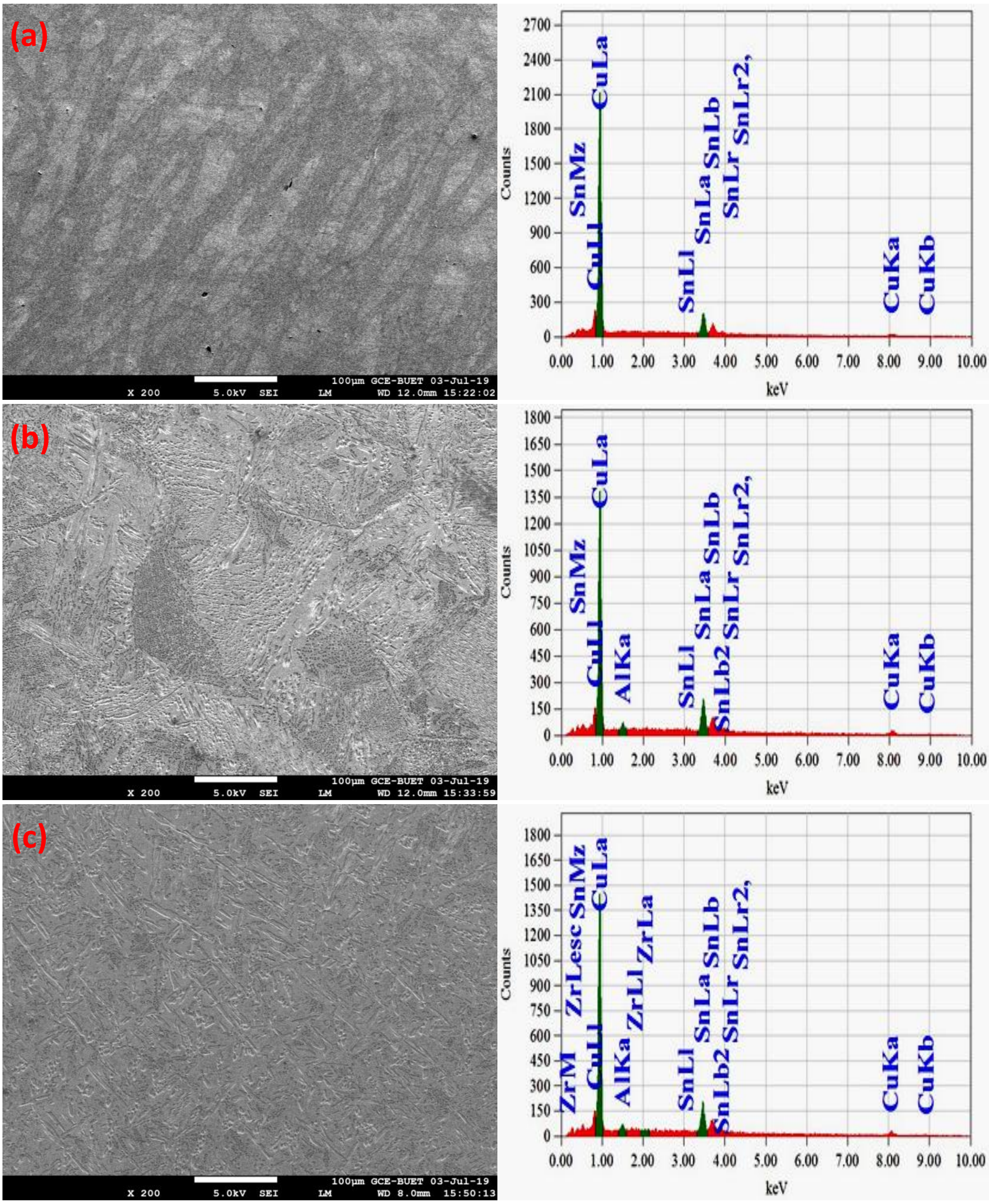

Figure 8. SEM images and and EDX analysis of the experimental alloys aged at 300°C for one hour a) Alloy 1, b) Alloy 2 and c) Alloy 3

4. Conclusions:

The ternary aluminium addition changed the physical properties of bell metal showing the ageing response by increasing the hardness of it while quaternary zirconium addition restricted the softening with the increase of temperature. The intermetallic precipitates of Al_2Cu, Al_4Cu_9 and Cu_3Al_2 formed in the ternary aluminium added alloys is the reason behind of this increasing hardness. This addition also changed the microstructure of the cast alloy where large dendritic arms formed and the quaternary addition of Zr refined the grain structure. Zr with a very trace addition also hindered the softening of the alloy due to the precipitation of Cu_9Zr_2 and Al_3Zr, which are immensely stable against grain coarsening, re-dissolution and pin-grain boundaries and thus increasing the thermal stability. Because of this, while the cast bell metal and aluminium added bell metal were observed to be completely recrystallized after ageing at 500°C for one hour, the alloy with trace amount of zirconium did not follow the same trend. The cast bell metal showed the best response in sound intensity level. This was because of the self-sustained oscillating ability with lower internal velocity and lower damping feature of the base alloy. However the quality of acoustic response of all three bell metal alloys was improved by age hardening. The quaternary Zr added alloy showed better acoustic response than ternary aluminium added alloy at differently ageing temperatures up to 500°C due to the grain refining ability of zirconium. Furthermore, the ternary addition of Al and quaternary addition of Zr affected the optical properties of the bell metal significantly. There was increase in the reflectance of the bell metal due to alloying additions. This variation of reflectance in the alloys was more prominent when they were aged at 500°C. As the grain size of base alloy and ternary Al added alloy increased at this temperature, the reflectance of the alloys were lower than that of quaternary Zr added alloy.

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