Effect of Aluminium Addition and Grain Refinement on the Microstructure, Mechanical and Physical Properties of Leaded Brass Alloys

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Abstract
Because Cu-Zn alloys have excellent combination of both physical and mechanical properties such as high thermal and electrical conductivities, good tribological behaviour, relative stability in corrosive environments and high mechanical strength, their study is very important for industrial and academic purposes. Review of previous experiments of leaded brasses focused on the effect of small addition of Al on microstructure and mechanical properties and did not evaluate physical behaviour of the alloy. In the current work, the influence of heavy aluminium addition and grain refinement for leaded brasses was investigated and the analysis is extended to physical properties evaluation. The results obtained show that mechanical properties are directly dependent on the microstructure which is in agreement with previous experiments. Initial addition of Al to the alloys does not result to heavy increase in hardness, but further increase in Al and addition of inoculants lead to significant change in hardness. The same pattern of modification was observed in both compressive strength and ultimate tensile strength. For the physical properties, there is little discrepancy to the sequence of their modification, most especially after grain refinement. There is a steady decrease in electrical conductivity even after grain refinement. This is not the case for thermal properties. The thermal conductivity reduced and then increased tremendously after Ti addition. The microstructure was modified with the addition of Al and Ti grain refiners. The presence of Al promoted formation of β-phase seen in the dark regions of the microstructure while Ti aided equiaxed grain formation.

Keywords: Brass, microstructure, mechanical properties, physical properties, equiaxed grains.

1 Introduction
Copper-Zinc alloys generally known as brasses have very wide range of applications such as musical instruments, decorative ornaments, industrial alloys and building services. The reason for the general acceptability for these alloys centred on their numerous excellent physical and mechanical properties. The compositions of zinc and copper can vary to create different types of brass alloys with varying mechanical and physical properties. Good electrical and thermal conductivities of brasses coupled with adequate mechanical strength make them versatile in electrical transmission, electronic devices, thunder protecting systems for telecommunications and buildings, electric motor and transformer installation applications. Their good colour reflection and excellent corrosion resistance attract them for ornamental and decorative purposes. High density and adequate mechanical strength of copper based alloys encourage their use for load bearing applications. Combination of workability and durability make copper alloys ideal for horns and bells while their good tribological behaviour outstand them for applications such as locks, gears, bearings, valves and ammunition casings. Initially, brasses are defined as Cu-Zn alloys with copper contents between 55-95 percent. Recent developments see these brasses alloyed with elements such as lead, aluminium, arsenic, nickel, cobalt, silicon, manganese, iron, tin, etc. to produce many complex wrought alloys with improved properties. Small additions of the alloying elements can largely influence the microstructure which also controls the mechanical and physical properties of the alloys. Relative low melting temperatures of copper brass alloys have made them attractive to primary manufacturers of nonferrous alloys due to low cost of production of billets, slabs and ingots. Also, their good malleability and machining properties make
them easy to be forged into different shapes by thermo-mechanical processing such as rolling, extrusion, wire drawing etc.

Grain refinement has been established to be one of the key phenomena to strengthen the properties of cast and wrought components. Brasses are susceptible to hot tearing and grain refining can remedy the situation to some extent. Maxwell and Hellawell [1] developed a simple mathematical model for grain refinement during solidification of alloys in order to elucidate the basic mechanism of nucleation and growth of dendrites. Lead is often added to Cu-Zn alloys to induce good machinability [2] that result in good malleability, softness and lubricity. The benefits introduced by the presence of lead have been appreciated for many years to facilitate chip fracture, reduce cutting forces, increase the machining rate and productivity, reduce tool wear, and enhance surface finish [3]. Lead also refine grain size but does not modify the morphology of the dendrites and insoluble eutectics. On the other hand, aluminium addition to Cu-Zn alloys confers hardness by enhancing the formation of β-phase and modification of grain morphology from dendritic to equiaxed structures [4]. Introduction of elements like Fe and Mn will lead to solid solution strengthening of copper based alloys by increasing the amount of hard β-phases, since they have poor heat treatment behaviour. Davies [5] observed that some alloying additions largely influence the structure of brass by altering the sequence of formation of alpha, beta or gamma phases present. When the proportion of these elements exceed some critical compositions, intermetallic or electron compounds may result instead of solid solution alloy thereby erasing the gains of structural modification and grain refinement. Therefore, brass alloys require to be strengthened by small amounts of additives to decrease the content of intermetallic compounds (IMCs) and disperse fine in the matrix [6]. Some analytical tools such as computer-aided cooling curve analysis (CA-CCA) [7] can be used to accurately predict the degree of grain refinement by calculating fraction of solid evolved during solidification as a result of melt undercooling and solidification time. Grain refiners are inoculants added to the solidifying melt to enhance heterogeneous nucleation and lead to formation of fine and uniform grain sizes and distribution [8]. These inoculants such as Ti and B provide substrates on which crystallites nucleate and grow leading to decreased grain size, hence result to improved room temperature yield strength, hardness, tensile strength, fatigue strength and impact strength [9].

Review of recent literature [10] shows that much work has been done to determine the influence of small addition of Al and grain refinement on the microstructure and mechanical properties of brass alloys. However, much has neither been said of the effect of heavy additions of Al on microstructure and mechanical properties nor extend the investigation on the physical properties such as thermal and electrical conductivities and the possibility of replacing large percentage of copper in brass with aluminium for cost effectiveness without adversely affecting their attractive properties, especially those that outstand them for electrical applications. This current research aims to close this gap through experimental analyses that would be described in subsequent sections. Titanium powder would be used for grain refinement for this work.

2. Methodology

The experiments are done in stages. Different grades of master alloys as samples were produced, prepared and then subjected to various tests ranging from microstructure, mechanical and physical properties analyses.

2.1 Sample Preparation
A crucible furnace for melting was made by positioning the vehicle wheel on the ground in a foundry workshop and a hole made on the side of the vehicle wheel. Six samples of different Cu-Zn-Pb base alloys code named \(A_n\), \(A_1\), \(A_2\), \(A_3\), \(A_4\), \(A_5\) with average weight of 0.35kg were produced through casting using high purity 99.99% copper. In the first alloy sample \(A_n\), Cu-Zn-Pb alloy was produced without addition of Al and Ti as modifiers and grain refiners respectively. The mixture was stirred thoroughly and continually during the melting to facilitate melt homogeneity and when the mixture was liquid enough (completely molten), it was then poured into the metallic mould and left to solidify. Subsequently, alloy samples of different percentages of Al were produced in similar method as the one described above. In the last sample \(A_5\), Ti was added to the alloy in order to induce grain refinement through rapid nucleation and growth processes. Extra 5% each of Zn and Pb were added to the melt to account for their loss through high temperature evaporation. Reduction of micro-gas porosities was ensured by the addition of degassing tablets. Small degree of superheating was carried out to reduce metal loss by evaporation and also to attain high fluidity which leads to improved castability of the alloys. Because Cu-Zn alloys are susceptible to hot tearing, the mould was preheated to 500°C to avoid sticking of molten alloy on mould surface and reducing the tendency of shrinkage induced cracks resulting from large thermal gradient between the melt and mould. Mass and chemical compositions of the various samples are given respectively in tables 1 and 2 below.

| Alloy code | \(A_n\) | \(A_1\) | \(A_2\) | \(A_3\) | \(A_4\) | \(A_5\) |
|------------|---------|---------|---------|---------|---------|---------|
| Cu         | 200     | 200     | 200     | 200     | 200     | 200     |
| Zn         | 70      | 70      | 70      | 70      | 70      | 70      |
| Pb         | 70      | 70      | 70      | 70      | 70      | 70      |
| Al         | -       | 15      | 20      | 25      | 30      | -       |
| Ti         | -       | -       | -       | -       | -       | 25      |

Table 1: Mass Composition of Samples (g)

| Elements | \(A_n\) | \(A_1\) | \(A_2\) | \(A_3\) | \(A_4\) | \(A_5\) |
|----------|---------|---------|---------|---------|---------|---------|
| Cu       | 58.56   | 56.31   | 55.39   | 54.42   | 53.96   | 52.41   |
| Zn       | 20.50   | 19.71   | 19.39   | 19.05   | 18.88   | 18.34   |

Table 2: Chemical compositions of Samples (wt %)
Microstructural Analysis
Microstructural test is one of the most important analytical tools for metallurgists and alloy manufacturers. Small pieces of disc shaped dimensionless specimens were cut out from the various samples for microstructural investigation. Standard procedures of sample mounting, grinding, polishing and etching for micrographic examination were ensured to obtain good details of the microstructure. A thermosetting phenolic powder was used to embed the sample with a hydraulic mounting press. The samples were manually ground with silicon carbide papers in the descending order of 220, 320, 500, 800 and 1000 grit sizes until very smooth surface is obtained. The surfaces of the samples were then polished on rotary polishing wheels with Diamond suspension and cleaned with alcohol.

![Fig. 1: Rotation of 90° on each successive grinding paper.](image)

The samples were etched with Ferric Nitrate and Hydrochloric acids to expose the grain constituents and grain boundaries which were viewed under metallurgical microscope and SEM.

2.3 Mechanical Tests
2.3.1 Tensile Test
The samples for tensile tests were prepared according to the standard dimension as shown in the fig. 2 above. Both ends of the samples were fixed to the chucks in the tensometer and load was applied until necking and breaking of the materials to determine the tensile behaviours of the original and altered alloys.

2.3.2 Hardness Test
Small pieces of non-dimensionalized specimens were cut out from the samples for hardness test. The test was carried out using Vicker’s hardness tester.

2.3.3 Compressive Test
Compressive test was carried out on machined samples with diameter of 20mm and height of 25mm, see figure 3 above. The samples were then tested using a universal testing machine and the maximum compression loads taken.

2.4 Physical property Tests
2.4.1 Thermal Conductivity Test
A heat conducting compound or heat sink was first applied on the surface of the samples to prevent air from passing through the gap where the sample is on the apparatus. The samples were then tested using a P.A. Hilton Heat Conduction Unit having a heater unit and a cooler unit. The samples were heated for 30 minutes each to ensure that the materials get saturated with enough heat to allow for complete isothermal transformation which helps to get a reliable result. A constant and continuous power supply of 20 watts was used. The thermal conductivity $k$, is calculated using the equation,
\[ k = \frac{Q}{A} \times \frac{dx}{dT} \] ........................ (1)

Where \( Q \) is power heat, \( A \) is the cross sectional area, \( dT \) is the thermal gradient while \( dx \) is the thickness of the material.

### 2.4.2 Electrical Conductivity Test

The test was carried out with rectangular samples cut-off from the specimen with no dimensions. The surfaces of the specimen was smoothened and cleaned to remove insulating substances that might introduce error in the result. Then, the samples were tested using the Magger Electrical equipment. The generator was hand cranked to produce a high direct current/voltage which passes a small current through the sample and the conductivity readings were taken. The electrical conductivity is calculated using the equation,

\[ \sigma = \frac{1}{\rho} \times 10Mho \] ........................ (2)

Where is the electrical conductivity, \( \rho \) is the electrical resistivity.

### 3. Results and Discussion

#### 3.1 Microstructural Investigation

Although, thermal conditions and solidification time have great influence on the microstructure that develop during solidification of alloys, but that is not the focus of this research. All the microstructures developed here are studied relative to effect of alloying additions and grain refinement on mechanical and physical properties of various brass alloys.

![Micrograph of the control alloy, A_n.](image)

The control alloy, code named \( A_n \), that is a mixture of Cu-Zn-Pb, was the first alloy to be cast whose optical micrograph is shown in figure 5, describes a typical microstructure of as-cast components without any modification or grain refinement. Without making reference to the cooling conditions, the micrograph presented above indicated that Cu-Zn-Pb system contains coarse grains with dendritic morphology. It is a confirmation of competitive growth between different phases, namely primary \( \alpha \) grains and \( \beta \)-phases. The primary \( \alpha \) dendrites are represented by the brighter regions while the second \( \beta \)-phases are represented in the dark regions as indicated by the arrows in figure 5 above. The beta phase can comprise insoluble eutectics, grain boundaries or particles of intermetallic or electron compounds.

Subsequent addition of other elements such like Al and Ti further modified the structure in terms of grain morphology and solid fraction evolution as a result of imposed solutal and curvature undercooling.
Since Al is β-phase former, it enhances nucleation and growth of second β-phase as a result it strengthened the mechanical properties of the alloy. The micrograph is shown in figure 6, more dark regions are noticed. Further additions of aluminium resulted to reduction in grain size as shown in figures 7 and 8.

Even though it did not cause much change in mechanical properties, it adversely affected the physical properties especially the electrical conductivity as would be seen in subsequent results. Fortunately, the decrease in physical properties is still far away from been detrimental, as the alloy can comfortably compete for thermo-physical applications without loss of efficiency.
Other β-phase formers include Fe and B, therefore, there is serious need to control the amount of these elements in copper brasses to eliminate the tendency for the formation of excessive intermetallic phases. This is because the presence of too much of these phases is capable of reversing the gains of solid solution strengthening. The ultimate grain refinement was achieved when Ti powder was introduced into the alloy, sample A5 as shown in figure 9 above.

3.2 Mechanical Test Analysis

The mechanical properties data of the investigated alloys are given in table 3 below. It can be seen that there is a slight decrease in hardness especially at low Al content A1, A1 and A2. But when the Al content is increased considerably as seen in A3 and A4 samples, there is a large increase in hardness of the materials. This can be correctly attributed to the formation of second β-phase and insoluble eutectics which harden the structure evolved since Al is a promoter of β-phase formation. Further increase in Fe and other alloying elements could lead to the formation of intermetallics and electron compounds, and therefore care must be taken to determine the quantity of alloying elements appropriate for a specific alloy. When inoculants (Ti powder) are introduced for the purpose of grain refinement, the polycrystalline equiaxed structure developed has positive influence on the ductility and hence leads to decrease in the hardness of the alloy. This is a typical condition observed in the sample A5.

Almost the same pattern of change is observed in their UTS and maximum compressive test analyses. There is no regular sequence of repeatability in decrease or increase of variable when aluminium is
been added. However, when grain refinement is carried out, a massive increase in both UTS and compressive strength is recorded as observed in Table 3.

### 3.3 Physical properties Analyses
Thermal and electrical behaviours are the physical properties considered for examinations in these alloys due to their frequent applications involving thermal and electrical stability. Hence, other physical properties like magnetic and optical behaviours are not examined because the applicability of these alloys in such fields can be considered as insignificant.

Table 3: Mechanical behaviour of the investigated alloys

| Alloy Sample | Hardness (Hv) | Ultimate Tensile strength, UTS (MPa) | Max. Compressive Strength (KN) |
|--------------|---------------|-------------------------------------|--------------------------------|
| A₀           | 45.9          | 95.54                               | 295.316                        |
| A₁           | 44.1          | 93.60                               | 101.83                         |
| A₂           | 43.7          | 102.71                              | 91.65                          |
| A₃           | 49.2          | 90.20                               | 86.558                         |
| A₄           | 53.6          | 88.6                                | 86.558                         |
| A₅           | 47.7          | 99.37                               | 142.566                        |

As expected, the addition of Al to brass alloys has a reducing effect on the electrical conductivity of brass alloys, as can be seen in Table 4 above. Even grain refinement was not able to circumvent this situation, hence further reduction in electrical conductivity was recorded in the refined alloy, A₅. Interestingly, the magnitude of the decrease in electrical conductivity with increase of Al content cannot be said to be detrimental to so many electrical applications, and brasses with high Al content is encouraged in such applications for cost effectiveness since aluminium is abundantly available and cheaper than copper. However, there are few special applications where their use cannot guarantee high efficiency. Heavily Al alloyed Cu-Zn brasses can be avoided in such applications. Somewhat different scenario is observed in the thermal conductivity behaviour. There is more irregular pattern in the increase and decrease in the thermal properties of the alloy as the amount of Al content increases. This is shown in the table displaying physical properties data above. There is a deviation from the electrical behaviour when grain refinement was carried out, where further decrease was observed. In the case of thermal property, grain refinement recorded jump in thermal conductivity, 113.489 KW/m.K for the grain refined alloy, A₅, as exclusively obtained in Table 4.

Table 4: Physical properties data obtained

| Alloy Sample | Electrical Conductivity (Mho) | Thermal Conductivity (KW/m.K) |
|--------------|-------------------------------|-------------------------------|
| A₀           | 100                           | 81.278                        |
| A₁           | 90.9                          | 32.256                        |
| A₂           | 76.9                          | 68.196                        |
| A₃           | 75.8                          | 54.271                        |
| A₄           | 74.1                          | 77.188                        |
4. Conclusion

The effect of Aluminium addition and grain refinement on the microstructure, mechanical and physical properties of leaded brass alloy was studied and the following conclusions can be drawn from the results obtained.

1. The presence of Al in the Cu-Zn-Pb alloy promoted the formation of second β-phase and affected the morphology as can be seen from the micrographs.
2. Addition of Ti as inoculants effectively refined the grain, reduces the dendrite sizes and increases the volumetric grain density.
3. The presence of the second β-phase, modified and refined grains affected both the mechanical and physical properties of the alloys, see tables 1 and 2 respectively.
4. The initial change in properties was not regular and heavy until grain refinement was achieved.
5. Addition of alloying elements decreased the electrical conductivity of the alloys progressively.

5. Recommendation

Since the presence of aluminium promoted the formation of second insoluble eutectic phases and led to the refinement of the grains in the alloy, which largely influenced mechanical properties of the leaded brass positively, it is recommended that more aluminium should be added to brass alloys. This will surely cause reduction in the prices of brass alloys since aluminium is more abundant than copper. However, care must be taken not to exceed 8%Al during manufacturing of brass alloys, especially those for electrical applications in order not to reduce performance of such components.

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