



Effect of 3 wt% Al Content and Rapid Solidification on the Erosion-corrosion Behavior of 92 wt% Cu and 5 wt% Ni Alloy in Seawater

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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ABSTRACT

A 92 wt% Cu and 5wt% Ni alloy containing 3 wt% Al is subjected to a rapid solidification processing (RSP) using a rotating copper wheel (7.95 cm ϕ x 5.95 cm w) at 1420 r.p.m, which produces a thin specimen of 2 mm thickness. The specimens with dimensions of 10.8x15.02 mm² are then exposed to water flow rate for 28 hours at 30°C and then the corrosion/erosion rates were computed in (g cm⁻² h⁻¹) using weight loss measurement method. The corrosion rate of the produced alloy under constant water flow rate tests decreases generally with time (1.5 - 28 h) and showed only initially some short periods of accelerated oxidation (0.0 - 1.5 h). The obvious weight loss and corrosion rate versus exposure time relationships are rationalized in terms of film formation was hard and its breakdown mechanisms did not occur. The effective of Al₂O₃ in improving the corrosion/erosion resistance was provided through addition 3% Al to the binary 92% Cu-5% Ni alloy. Thus, a passivity, dense, adherent, and continuous Al₂O₃ film exists on the surface protecting bare alloy from further oxidation and causing the metal to be little deterioration Moreover, fine inner layer of Cu₂O or CuO acts as a super resistance barrier to erosion-corrosion. The

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metallographic images were photographed using a metallurgical microscope at 1000X magnification to identify the influence of rapid solidification on the structure includes nucleation, grain size, grain growth and particles pushing.

Keywords: 92Cu-5Ni alloy; rapid solidification; erosion-corrosion; cupronickel alloy.

1. INTRODUCTION

Because of the importance of the cupronickel alloy and its various application in our industrial life, addition to that their use in human body as implantation, medical micro devices and high technology there are needs to develop a new alloy posses higher properties than that available. If the science can develop one has an improvement only of 2%, the mankind becomes in wealth on all aspects.

Cupronickel alloys (10-30% wt Ni) are widely used in condensers, condenser plates, distiller tubing, desalination plants, navies, water cooled, heat exchangers, hydraulic pipeline, etc. due to excellent impingement corrosion, stress corrosion cracking, and uniform and localized corrosion resistance in seawater. The 70/30 cupronickel ally show better resistance than 90 Cu-10 Ni alloy to erosion- corrosion up to 3.5 m.s^{-1} [1-8]. Copper base alloys containing 10-25 wt % Ni promotes resistance to impingement in both fresh and salt waters and to stress corrosion cracking [9].

However, the alloys composed only of Cu and Ni neither has adequate strength nor sufficient resistance to corrosion to serve in sea water and off shore. At the metallurgical properties, iron content and microstructure have been reported to play major roles in affecting the corrosion performance of cupronickel alloys in seawater. Iron acts as a grain refiner and increases tensile strength. Copper-nickel contains 1% to 4% Fe and 5 to 10% wt Al are added to improve erosion-corrosion resistance at seawater velocity from 1.5 to about 4.5 m/s [10-11].

Besides the additives that are added, severe segregation, coarse and heterogeneous casting microstructures, undesirable massive grain boundaries occur during conventional slow solidification process i.e. ingot or mould casting [12-14]. Erosion-corrosion is commonly observed at elbows, bends and tees piping. In piping systems, erosion-corrosion can be reduced by increasing the diameter of the pipe, thus decreasing velocity and turbulence of fluid flow. Inlet pipes should not be directed onto the vessel

walls if this can be avoided [15-19]. Several rapid solidification techniques to fabricate rapidly solidified alloys (at cooling rates of 105-107°C/sec) as ribbons, filaments, wire, flakes, or powder in large quantities have been investigated to develop new copper-nickel alloys. One well known example is melt-spin chill casting, whereby the melt is spread as thin layer on a conductive metallic substrate moving surface to produce continuous ribbons or tapes.

Rapid solidification process has been shown as a powerful means for achieving higher solubility of alloyed elements in solid solutions, what is especially important of a production of alloys with elements that have small solubility in equilibrium state with unique structures and properties [20-22]. In addition, they exhibit excellent electrical conductivity, complete chemical homogeneity and mechanical characteristics quite different from their slowly cooled counterparts moreover. Furthermore, melts undercooling opens new solidification pathways for new non-equilibrium phases and non-conventional microstructures as structural refinement, new metastable crystalline, novel crystalline or amorphous and solid solubility extension in the form of fine dispersoids, rather than segregated particles [23-25].

Erosion–corrosion is known to promote the premature failure of materials when exposed to erodent in a corrosive environment. The combined effects of erosion and corrosion can be significantly higher than the sum of the effects of the processes acting separately. Thus, there remains a need to develop a new copper base alloy with homogenous chemical composition and structure securing more stable erosion-corrosion resistance for numerous engineering applications.

The focus of the article is to investigate the effect of rapid solidification and 3% Al addition on the erosion-corrosion behavior on 92% Cu-5% Ni alloy in water. The work includes preparing materials and designing an apparatus for producing thin specimens, which were then subjected to erosion-corrosion operation tests.

2. EXPERIMENTAL

2.1 Materials

The starting materials for all ingots were minimum 99.9% pure, except the copper that was 99.999% pure. The sample of this alloy was supplied in the form of ships. The ships of Cu/Ni were machined by a lathe and Al ships by a drill machine to fill the clay-graphite crucible.

2.2 Rapid Solidification Device

(Fig. 2.1) shows the apparatus which was designed by the present author in order to accelerate solidification of the 92% Cu-5% Ni alloy containing 3% Al. This apparatus was a rotating copper wheel (7.95 cm ϕ x 5.95 cm w), which was connected with stainless steel shift, a motor gives a rotating velocity of 1420 r.p.m, and crucible holder punched in its bottom (2 mm ϕ), and connected with a nozzle throughout its bottom orifice. This entire component fixed to a frame in carbon steel table to avoid vibration of the flowing molten alloy through the nozzle downward onto rotating copper wheel surface, which conduct the latent heat away from the melt to produce a continuous thin plate of several millimeter in thick.

The horizontal distance between the nozzle and the rotating wheel was 4 cm. The temperature conditions vary with the mixing amount of the alloying elements. The required amounts of copper, aluminum, and nickel were weighed in a digital balance. Since the melting point of Al metal is lower than that of the copper base alloy, Al ships are enveloped within the aluminum voile in order to prevent the loss of Al in the prepared alloy by its vaporization during melting process. The Ni, Cu, and Al ships were added to a crucible within an electric furnace (1060°C) for 3 h under a protective environment, withdrawn the crucible contained melted alloy via ladle at relatively high speed, and poured in the cylindrical graphite crucible pored at the bottom that was connected with nozzle (0.3 mm Φ) that, which molten alloy is flew downwardly on a conductive rotating copper wheel surface with Φ 7.95 cm and velocity/speed of 1420 r.p.m whereby the melt is spread as a thin layer to form a specimen of 2 mm in thick.

The containing molten alloy crucible with a pored bottom diameter of 2 mm was placed 3 cm on the top of the rotating copper wheel i.e., the distance between nozzle and rotating wheel was

3 cm. The width and thickness of the thin specimen depend on the wheel speed, crucible orifice diameter, height of the orifice to the wheel. The wheel speed mentioned in the text is the linear speed which is circumference of the wheel (in m) times the rotational speed (in revolution per second).

2.3 Erosion-corrosion Test

2.3.1 Erosion-corrosion device

The erosion-corrosion tests of rapid solidified samples were carried out using a self-designed erosion testing device which includes two open tanks of 25 cm height x 50 cm width x 50 cm length as shown in Fig. 2.2. A first tank being mounted on a table and a second tank being mounted on the ground in which the vertical distance between them was 40 cm. Specimen with exposure area of 162.1737 mm² was fixed on a holder positioned in the center of an upper tank. Inlet and outlet tubes were connected to water pump of 0.75 hp to pump the water to the upper tank through copper tubes which then flow down into ground tank due to elevation. Water flow rate was directed against the surface area of the specimen.

2.3.2 Erosion-corrosion specimens

The specimens of the 92% Cu - 5% Ni alloy containing 3% Al were cut out in dimensions of approximately 10.8 x 15.02 x 3 mm³ for erosion-corrosion testing, and then cleaned in sequence with emery paper of different grades, polished, washed with distilled water, and acetone and dried. The specimen was fixed on the middle of copper rod by locking arrangement and mounted in a non-conducting epoxy resin with the 10.8 x 15.02 mm² face areas being exposed to the test water flow rate (7.5 pH), open to air at 30°C.

The facing surface area of the specimen was exposed to 270.4 kg/min water velocity impact in a perpendicular direction for 28 hours. The duration time test was started from zero time and stayed for 30 minutes, and then duration time test was increased by 30 minutes step by step until to 28 hours with unchanged in flow rate.

2.3.3 Erosion-corrosion evaluation

The experiments were designed focusing on the effects of aluminum addition and rapid solidification of Cu-Ni alloy on erosion/corrosion performance. Before and after testing, the

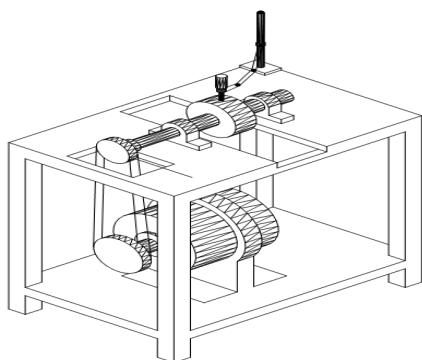


Fig. 2.1. Rapid solidification device

samples were cleaned with acetone, degreased, rinsed, dried and weighted using an analytical balance with an accuracy of 0.0001 mg. The erosion and /or corrosion rates were calculated by weight loss methods. Measuring the weight change per unit area and the extent of erosion–corrosion of the specimens assessed the erosion–corrosion behaviour of the alloy.

The weight change consists of weight gain owing to the formation of oxide scale and weight loss due to the erosion. The net weight change represents the combined effects of these two processes. The weight loss is the weight difference in gram per square centimeter (g cm^{-2}) between the initial weight of the specimen before the test and that of the specimen after being impinged with the water flow rate for certain time in hours. The scale thickness lost due to the erosion was assessed by finding the difference in thickness of the specimens before and after 28 hours exposure to water flow velocity impact. The thickness can be measured with a micrometer or ultrasonic probe test. Specimens with different thicknesses can be obtained by varying the cooling rate due to varying rotating copper wheel speed. At lower rotating wheel speeds, a mixed structure of mostly amorphous regions and small fraction of crystalline phases. At higher wheel speeds a completely amorphous on substrate as well as on air-cooled side can be obtained [26]. The corrosion and or erosion rate can be computed using weight loss of the specimen and the following equation will be employed:

$$C.R = \frac{(W_1 - W_2)}{A t} \quad (1)$$

where W_1 and W_2 are the initial and final mass (g) of the sample before and after exposing for a

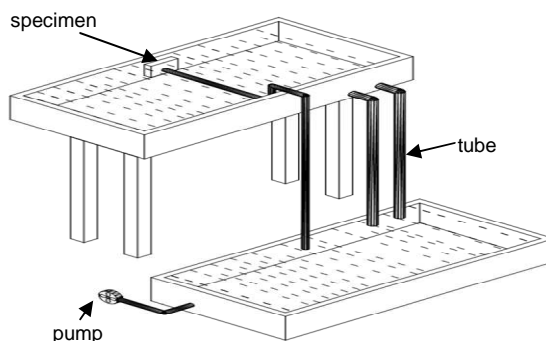


Fig. 2.2. Corrosion/erosion device

specific time , A is the exposure surface area of the specimen (cm^2) and t is the certain interval time (h).

W_1 = original specimen mass (194.998×10^{-3} g);
 W_2 = final specimen (g); A = Specimen Area (1.6217 cm^2).

2.4 Microstructure

The specimens were finely polished with different abrasives paper grades (320, 500, and 1000), etched, and dried. The metallographic images were photographed using a metallurgical microscope at 1000X magnification. Due to similar erosion–corrosion behaviour of all the specimens, analysis of only representative micrographs is presented here.

3. RESULTS AND DISCUSSION

3.1 Erosion–corrosion Rate

The designer of materials to be exposed to water flow velocity is dependent on the circumstances operating environment, ability of the material to withstand erosion-corrosion attack, which is combined action of corrosion and erosion that removes any inherent or applied protective film, causing localized material loss; and the effects of erosion-corrosion impose economic constraints in many industrial applications. It occurs in areas where the effects of water flow rates or steam impingement on the alloy surface are great enough to cause mechanical or electrochemical disruption of the protective oxide film.

Corrosion rates can lead to loss of wall thickness and to high release rates of material into the flow stream. This form of attack depends on

hardness, surface smoothness, size and distribution of particles, structure, flow velocity, bubble size, angle of incident, turbulence and the formation of a protective film and its ability to re-passivation and thickening of the surface film.

The variation of erosive-corrosive rate of 92% Cu and 5% Ni alloy as a function of time in sea water is shown in Figs. 3.1 and 3.2. Weight loss increases rapidly for the first 12 hours, and then it keeps constant until the end of the test at 28 hours. This is may be attributed to weakness of the film formed of alumina on the surface of alloy produced, and after that the film becomes dense, adherent, and continuous Al_2O_3 film exists on the surface protecting bare alloy from further oxidation and causing slow down the corrosion rate. Initially, for the first 15 min, the weight loss and corrosion rate were remind unchanged. This may due to the formation of a slim film of Al_2O_3 on the surface during specimen preparations. After 15 minutes, especially at interval time of 30 min, the specimen began to show an increase in the weight loss ($2.4665 \times 10^{-7} \text{ g cm}^{-2}$) and corrosion rate ($4.9329 \times 10^{-7} \text{ g cm}^{-2} \text{ h}^{-1}$). The water velocity impact led to remove the initial Al_2O_3 formed on the surface during specimen permitting further Al_2O_3 to take place at that specimen surface thereby increasing the weight loss.

It is clear from Fig. 3.1 that the percentage weight of Al not only affects the initial weight loss, but also affects the range of corrosion rate increasing over the time. The weight change per unit area (g cm^{-2}) verses time plot for 28 h exposure of the alloy to the water flow velocity is shown in Fig. 3.2. There is a small rise of

corrosion rate within the first 30 minutes and then remaining nearly the same. This demonstrates that protective Al_2O_3 layer formed on the surface of as-casted alloy still keeps stable with the increase of exposure time to 45 minutes. On other hand, the weight loss is reached $3.6998 \times 10^{-7} \text{ g cm}^{-2}$ and this means that the erosion attack still continued causing the removal of the corrosion product, which exposes a new reactive area to the corrosive influence.

Weight loss and corrosion rate keep increasing sharply until breakdown (1.5 h), but decrease thereafter. Such phenomena can be explained in term of the erosion-corrosion, since the passive oxide layer is broken and the underlying alloy is corroded, showing an increase in the erosion; there exists a competitive process between the passive film thinning and removal by mechanical erosion and self healing of passive film. When the rate of re-passivation cannot catch up with the damage induced by erosive action, consequently a significantly increase of corrosion rate happens.

Within the period of 1.5- 2 h the weight loss and corrosion rate were increased from $9.8659 \times 10^{-7} \text{ g.cm}^{-2}$ and $6.5773 \times 10^{-7} \text{ g cm}^{-2} \text{ h}^{-1}$ to $11.7158 \times 10^{-7} \text{ g cm}^{-2}$ and $5.8579 \times 10^{-7} \text{ g cm}^{-2} \text{ h}^{-1}$. Within the first 1.5 h the corrosion rate is high exhibited $6.5773 \times 10^{-7} \text{ g cm}^{-2} \text{ h}^{-1}$. The increase in weight loss appeared to be associated with the removal of oxide film and a continued effect of erosion, which prevents permanent formation of a protective film, and the corrosion of bare alloy consequently exposed to water flow velocity that can lead to rapid surface attack causing alloy loss and often penetration.

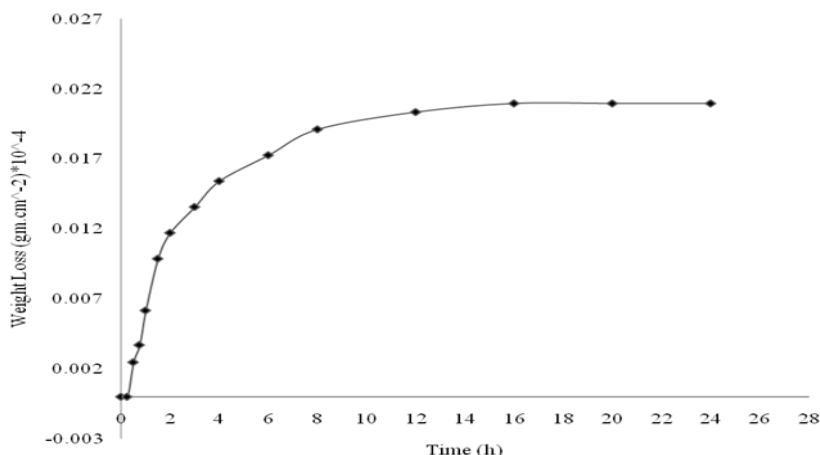


Fig. 3.1. Variation of the weight loss ($\text{g cm}^{-2} \times 10^{-4}$) with exposure time for 92% Cu-5% Ni alloy containing 3% Al after a testing period

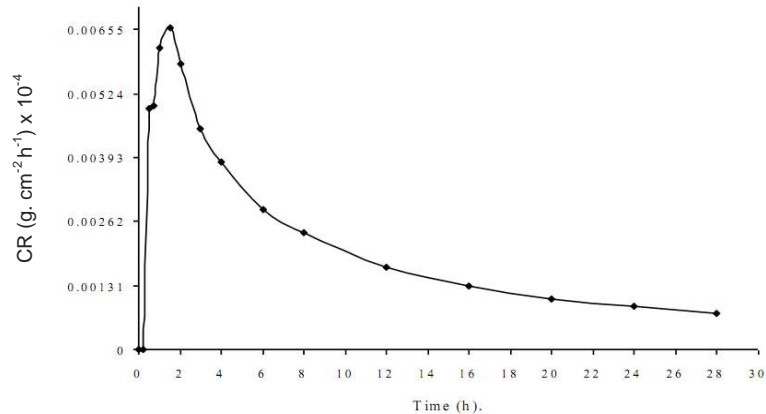


Fig. 3.2. CR (g cm⁻² h⁻¹) x 10⁻⁴ against time of rapid solidified 92% Cu-5% Ni-3% Al alloy in sea water for 28 hours at ambient temperature

3.2 X-ray Diffraction

The X-ray diffraction pattern of the Cu-Ni-Al alloy showed only DO type peaks, but a metallurgical micrograph indicated that numerous fine grains were dispersed in each grain with the size of several microns as shown in Fig. 3.3a. The results suggest that the rapid solidification rate at high temperature which resulted in the grain reinforcement of the Cu-Ni-Al alloy.

3.3 Microstructure of Rapid Solidified Cu-Al-Ni Alloy

The microstructure of cast Cu-Ni-Al alloy after 28 hours of exposure to water flow rate is shown in Fig. 3.3b. On the metallographic images thus obtained it was found that the grain size of the cast alloy is finer due to no further grain growth of the particles can occur upon contact with rotating wheel surface since the rate of cooling depends on thickness of cast specimens.

The difficulty in the growth of small particles in a large size on rotating copper wheel is due to insufficient time available for grain growth. The influence of rapid solidification on the structure includes nucleation, grain size, growth, and particles pushing. Such phenomena as described depend on cooling rate, i.e., surface quality of the wheel, heat. The effect of aluminum content on the erosion-corrosion properties of 92% Cu-5% Ni alloy also has been linked to the characteristics of the passive film. It has been suggested that the presence of aluminum in 92% Cu-5% Ni alloy promotes the formation of a film, consisting of corrosion product containing aluminum oxide that is effective in reducing the corrosion rate of cupronickel alloys.

This improvement of corrosion resistance of alloy due to the passive film of alumina, as well as it is attributed to the Al with +3 valence forms Al₂O₃ or Al(OH)₃ in water, which leads to strengthening of the alloy. Since the hardness of the specimens increased, in spite of damaging the oxide film formed on the sample surface. It has been proved that the passive film formed on the Cu-based alloying layer was mainly consisted of Al₂O₃, which ensured the highest erosion-corrosion resistance in rapid solidified for this alloy.

Although both Cu and Ni have face centered cubic (FCC) crystal structure but when both are mixed in any proportions, they form a solid solution having FCC crystal structure with a single phased (α) at room temperature [27]. Furthermore, the thin specimen has a single phase (α) microstructure. It is evident from the Fig. 3.3 that, the surface area of specimen of 92% Cu-5% Ni- 3% Al illustrating the fine grain size and uniform in size, and is distributed evenly. This result is consistent with the published information, that when a proper amount of Ni and Al is added together to Cu, the grains are made smaller and the length of the phase (α) becomes shorter as the grains are dispersed finer and more uniform, all of which contribute to improve erosion-corrosion resistance of copper base alloy.

As the grains are dispersed finer and more uniform, the length of the phase becomes shorter, while the surface condition is poor when the grains are coarsened. No casting defect is obtainable since dendrite arm is not formed, the surface is smooth, and the micro-segregation removes altogether and significantly reduces

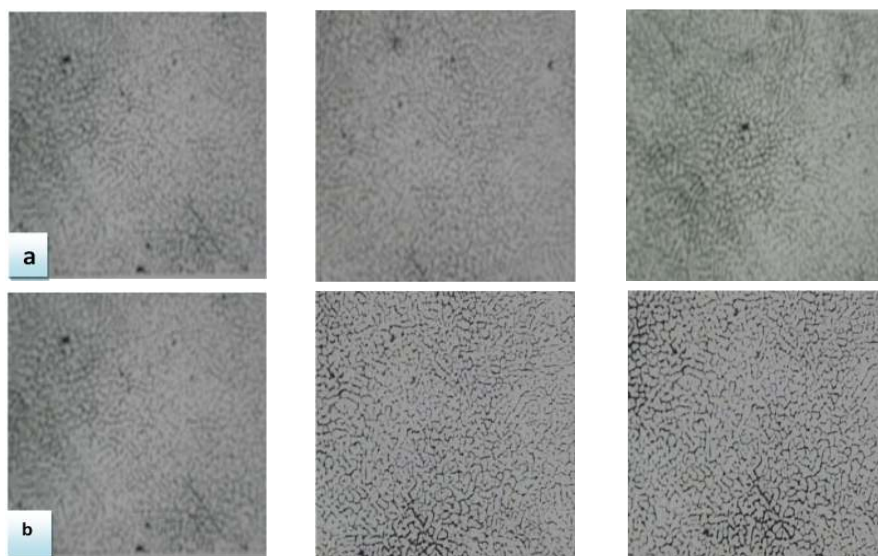


Fig. 3.3a and b. The microstructure of erosion-corrosion of a rapid solidified 92% Cu-5% Ni-3% Al alloy after 28 hours exposure to flow rate of normal water at ambient conditions

spacing over which micro-segregation. In contrast with counterpart alloy made while the poor surface coarse grains, brittleness etc. This means that the cooling rate is quite rapid to allow this phase to occur and any other phase is retained, i.e., a direct consequence of the grain refinement and a single phase microstructure achieved.

This result is consistent with the published information, that the addition of Al in less than 0.8wt%, to copper-nickel alloys leads to have identical single-phase (α) structures depending on the cooling rate [28]. Literature reported that the addition of aluminum to a Cu-Ni binary alloy increases the strength through the formation of nanometre-scale Ni_3Al (γ) phase precipitates [3,7,8]. An amorphous can be incorporated into the structure of an alloy by increasing the cooling rate during formation of thin ribbons. It would be expected, then, that the alloy would be excellent in strength and hardness, and also in heat resistance [6]. Some researchers [18,29] have found that the added nano-particulates improved the corrosion resistance.

4. CONCLUSIONS

The corrosion-erosion behavior of ternary Ni- Cu - Al intermetallic alloys as possible materials to be used in industries environments has been evaluated. All results have shown that their corrosion- erosion resistance was lower than that for conventional 70/30 and 90/10 cupronickel alloys due to the formation of passive film on the specimen surface.

The increase in corrosion rate for 92% Cu-5% Ni alloy containing 3% Al was sharp at the beginning of the erosion-corrosion test, but it was slowing down with the time until about 1.5 hours where it kept approximately constant until the end of test at 28 hours. The results showed that the aluminum oxide start to be formed in a short time producing hard layer of Al_2O_3 on the specimen surface, causing an increase in hardness of the considered alloy, which is mainly contributed for better performance of the alloy in the given environment.

After erosion-corrosion test, the microstructure of the specimens were analyzed by metallurgical microscopically examinations at 1000X magnification. Rapid solidification has a benefit that the grain growth was fine due to there was not sufficient time to for more growth. Therefore this alloy could be the promising material for the mineral industries where the components are subjected to severe erosion and corrosion in abnormal environments.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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