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Optimization and Modeling of Oxidation Process Parameters of α-Brass Alloy

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ANSTRACT: The objective of this work is to optimize the parameters (time and temperature) of the oxidation process and the alloy composition (alloy type) with addition of a small amount of oxide particles (0.2%TiO_2, 0.2%Y_2O_3 and 0.2%Al_2O_3) separately to **α**-brass alloy. Taguchi optimization with L16 orthogonal array, signal to noise ratio(S/N) and analysis of variance (ANOVA) were applied on these four alloys. In this study, the weight gain per unit surface area (ΔW/Ao) of the oxidized specimens for characterizing the oxidation extent was measured at different oxidation temperatures (500, 600, 700 and 800°C) and times (1, 10, 30 and 50) hrs for the base alloy and three modified alloys containing a small amount of oxide particles of 0.2%TiO_2, 0.2%Y_2O_3 and 0.2%Al_2O_3, respectively. It was found that the weight gain/surface area (ΔW/Ao) increases continuously with increasing time and temperature for the investigated alloys. It was obtained that the alloy containing 0.2% Al_2O_3 has the lowest rate of oxidation and reveals great enhancements in the oxidation resistance in comparison with the base alloy at all temperatures. It was found that the contribution percentage was 33.78% for oxidation time followed by oxidation temperature 30.9% and alloy type (modified brass) 19.94%. The results of the weight gain (ΔW/Ao) measurement propose that the oxidation kinetic of the whole investigated brass alloys follows the parabolic law in most experimental tests under various temperatures.

KEYWORDS: Alpha brass alloy, Oxidation process, Oxide particles, Taguchi method, S/N ratio, ANOVA

INTRODUCTION

Copper (Cu) and its alloys are broadly utilized in a diversity of products that assist and improve everyday lives. They possess excellent thermal and electrical conductivities, display a good formability and strength, possess the exceptional corrosion and fatigue resistance, and are in general non-magnetic. They are easily brazed and soldered, and numerous of them are welded via different resistance, arc, and gas techniques.

Copper and specific bronzes, brasses, and copper – nickel alloys are utilized widely for the heat exchangers, automotive radiators, solar collectors, home heating systems, and different other uses that need fast heat conduction across or alongside a section of metal. Also, due to of their exceptional capability to resist the corrosion, coppers, bronzes, brasses, and copper-nickel alloys are utilized for valves, pipes, and fittings in the systems that carry drinking water, treatment water or the other aqueous fluids, and the industrial gases [1]

All metals tend to form oxides in air at the room temperature despite in numerous examples the reaction rate is too sluggish at the low temperatures. The of rates reaction raise quickly with every temperature rise, and at the elevated temperatures, the majority of reactions are accomplished in a few minutes. Thus, relying upon the stable circumstances, the reactions of O_2 with the metal are nearly probably to take place, and the problems of oxidation are frequently decreased to determine the rate of reaction. Essentially, the oxidation at elevated temperature can be separated into (2) restricted types of processes: (1) Processes of oxidation, in which the atoms of O_2 don't melt in the base metals; (2) Processes of oxidation, in which the base metal depicts the O_2 solubility [2, 3].

The oxidation initiates with the O_2 dissolution into the layer of surface. If the O_2 concentration in the layer of surface attains the balanced concentration for the less-noble alloying element oxidation, reaction takes place and the solute oxide initiates starts growing on surface. The likely shapes and mechanisms of the growing of layer are similar to the initial situation. At a separate location, the O_2 concentration raises during the time and, once more, if the balanced concentration to oxidize the solute element is determined, reaction will take place and oxide the particles, Such operation possesses the term "internal oxidation", and the layer that comprised the precipitated oxide particles within the matrix is named "subsurface" or "zone of internal oxidation" [4, 5].

Taguchi techniques are statistical approaches established viay Genichi Taguchi for improving the produced goods quality, and in recent times, they are also employed to the engineering (Rosa et al. 2009), biotechnology (Rao et al. 2008; Rao et al. 2004), as well as marketing and advertising [6-8], correspondingly. Expert statisticians have greeted the aims and enhancements fetched around via Taguchi techniques, especially via Taguchi's designs evolution to study the change. Designs of Taguchi offer a strong and effective technique to design the processes that work steadily and optimally over a diversity of circumstances. For determining the best design, it needs the usage of a tactically designed experiment that subjects the process to different design parameters levels.

Certain too informative investigations were performed employing the Taguchi parameter design technique to optimize the parameters of oxidation process at the elevated high temperature uses. Each utilized different combinations and levels of temperature, time and alloy composition. Such investigations explored obvious and beneficial correlations between their parameters of response and control. That would reveal there is a no. of various parameters, which can be comprised in such kind of investigation, and a single parameters combination can be designed to fit a certain case [9].

Muna et al. [10] (2015) studied the (Al-Li) base alloy oxidation when this alloy contained small quantities of rare earth oxides, like (0.2wt%Y_2O_3) and (0.2wt%Nd_2O_3) particles at the temperatures (400ºC, 500ºC and 550°C) for (60hr) in the desiccated air. Within such investigation, a Minitab software package (Taguchi design method) was employed for estimating the optimum parameter of the weight gain/area (ΔW/A) in the oxidation operation of the (Al-Li) base alloys for obtaining a least oxidation layer thickness. An experimental design of (L9) Taguchi's orthogonal array was utilized for determining signal to noise ratio (S/N). The analysis manifested that the influence of different factors of input upon (ΔW/A) in the order of its influence is temperature, the kind of alloy, and the time.

Limited researches studied the effect of alloying elements or oxide particles on the oxidation rate and the resistance of copper base alloys, especially α-brass alloys

Muna K. A. [11] in (1997) studied the effect of temperature and chemical composition on the oxidation resistance and the thermal shocks of copper-base alloy (Cu-1%Al) alloy, which is used in the electrical applications and condenser tubes. Many Cu-base alloys with the addition of a small amount of 0.1%Te (Tellurium) and 0.1% Y(Yttrium) in the (Cu-1%Al) alloy were prepared by melting and casting in a metallic mold. Oxidation and thermal shocks tests were carried out in air at different temperatures (300-800ºC) for (53 hr). The results showed that the Y-containing alloy gave the least rate of oxidation and high improvement in the oxidation and shock resistances in comparison with the other investigated Y-free alloys.

Muna K.A.[12] in (2000) improved the oxidation resistance of α-brass (70Cu/30Zn), which is used in the heat exchanger tubes in power stations. Many brass alloys with the addition of a small amount (0.25-1) wt% of pure aluminum to brass alloy were prepared by melting and casting in a metallic mold. Oxidation and thermal shocks tests were conducted in air at different temperatures (500-800°C) for (53 hr). The results elucidated that the brass alloy containing (1%Al) exhibited a higher oxidation resistance (lower weight gain/area) than other alloys.

The oxidation of copper and its alloys is a significant subject for the academic research and the industrial uses. Therefore, the copper oxidation behavior has taken a significant attention for a long period of long. At the temperatures over (600°C), it's thought that the oxidation is governed via the Cu ions lattice diffusion throughout a layer of (Cu_2O) [13-15]. Dissimilarly, the documented kinetic data upon oxidation less than (500°C) changed considerably. Zhu et al. [16] in (2006) achieved a kinetic study upon the Cu oxidation above a range of temperature from (350°C) to (1050°C). It was proposed suggested that the oxidation during a range of temperature from (350°C) to (550°C) is governed via the diffusion of grain boundary. Shao-Kuan Lee et al. [17] in (2016) investigated the Cu oxidation behavior at temperatures at (200°C) and (300°C) and its mechanism. The oxides that developed beyond the oxidation at low temperatures possess fine crystal sizes; the constants of oxidation rate attained ($2\times10^{-15} m^2/s$) and ($6\times10^{-14} m^2/s$) at (200°C) and (300°C), correspondingly. The passivation processing at (600°C) in N_2 made a thin layer of oxide comprised comparatively big crystals of Cu_2O. The existence of this layer decelerates down the constants of oxidation rate via a value order. Raising the size of crystal in the oxide of surface decreased the rate of oxidation considerably.

The aim of this study is to optimize the oxidation process parameters and improvement the oxidation resistance of α-brass alloy (70 Cu/ 30 Zn) by addition of a small amount of oxide particles (0.2%TiO_2, 0.2%Y_2O_3 and 0.2% Al_2O_3) separately. Oxidation process at different oxidation temperatures (500, 600, 700, and 800) °C in dry air and times (1, 10, 30 and 50 hr) are investigated. The optimization of oxidation process parameters was applied using Taguchi method L16 orthogonal array and design of experiment (DOE) to find the optimal parameters (time, temperature and alloy composition) depending on the type of the explored oxide particles Also, by using "Taguchi's method" of design of experiments, a mathematical model was developed using the oxidation process parameters of α-brass alloys, such as oxidation temperature, time and alloy composition or alloy type containing oxide particles. Beyond collecting the experimental data, signal-to-noise (S/N) ratios were computed and utilized for obtaining the optimum levels for each parameter of input.

EXPERIMENTAL WORK

Preparation of Specimens

Four alpha brass alloys were produced via melting process and pouring the molten alloy into a lasting steel mold. In this research, the oxides particles of (0.2%TiO_2 ,0.2%Y_2O_3 and 0.2%Al_2O_3) were individually introduced into the molten of brass alloy (70%Cu/ 30%Cu) by gravity die casting method using mechanical stirring to disperse those oxides particles in the molten base alloy and pouring the molten alloy into steel molds. Homogenization heat treatment at (550°C) for (3 hrs) was carried out in an electric furnace (Carbolite type) to obtain a homogenous structure and reduce the segregation of impurities in ingots

The specimens for oxidation were machined from the ingot beyond the homogenization in form of discs having dimensions of (15 mm) in diameter and (2-2.5mm) in thickness. Such specimens were first ground via utilizing emery paper (SiC) having various grits (220, 320, 500 and 1000), and after that a polishing operation was conducted via employing diamond paste (grade 1μm) with distinctive polishing cloth as well as lubricant. Then, they were washed by water and alcohol and desiccated by hot air. The specimens were oxidized at a (500°C-800°C) temperature range in the still air at the atmospheric pressure in a perpendicular tube furnace having a homogenous hot region. The hot region temperature was governed via a thermocouple kind (K). The kinetics of oxidation were obtained via the technique of weight gain/area with a microbalance having a (0.1 mg) accuracy, where the specimen was located into the hot furnace beyond attaining the needed temperature, each specimen was hung using pt-wire. The weight of specimen was recorded before and after the oxidation at a given temperature to calculate the weight gain per surface area for all brass alloys at different temperatures.

Plan of Experiments and Analysis of Data

In the present investigation, the analysis depended upon the technique of Taguchi was conducted by employing Minitab 16 software for estimating the important factors of the process parameters of oxidation and the graphical analysis of the resulted data. Taguchi's orthogonal array is the most efficient design, employed for estimating the principal effects utilizing only few experimental tests. Such designs can study the principal influences when the factors possess more than (2) levels. In Taguchi technique, the analysis variation is achieved utilizing Signal to Noise ratio (S/N). Three (S/N) ratio methods of familiar attention exist for optimization [18].

In the current study, the goal is the minimization of the parameter of the weight gain/area (ΔW/Ao) for producing a minimum oxidation layer thickness. Consequently, the (S/N) ratio for every experiment of (L16) (3 variables x 4 Levels) was computed utilizing the smaller-the better method. The aim of employing the (S/N) ratio as a measurement of performance is to evolve a product and process unresponsive to the noise factor. In Taguchi technique, the word "signal" denotes the wanted value (mean) for the characteristic of output, and the word "noise" denotes the unwanted value (square deviation) for the characteristic of output. Thus, the (S/N) ratio is the ratio of mean to square deviation.

The overcut (S/N) ratio is computed via:

$$\frac{S}{N} = -10 \log \left(\frac{(\sum Y_i)^2}{n}\right) \tag{1}$$

Where, Y_i: is the ith response observed value.

n: The observations number.

RESULTS AND DISCUSSION

Taguchi Method of Doe

The method of Taguchi is a powerful tool to design high quality regimes. For increasing the experimental efficiency, the table (L16) blended orthogonal in the quality design of Taguchi has been utilized for determining the important factors [9]. Within the experiments part of the present study the experiments part, six influential oxidation process parameter were selected, such as oxidation temperature for different alloy types depending on type of oxide particles added to the base brass alloy, (each of which possesses (4) various levels (high, medium and low levels).

Figure 1 depicts the principal influences of the weight gain/area (ΔW/Ao) of every factor for different level circumstances. Referring to such figure, the (ΔW/Ao) reduces with the low temperature level and low time level. It was shown that the Figure 1 indicates the main effect plot for the mean of the process parameters of oxidation (temperature, time and type of alloy, which contains each one of oxides (0.2%TiO_2, 0.2%Y_2O_3 and 0.2%Al_2O_3) upon the weight gain/area ratio (ΔW/Ao). The (S/N) ratio analysis of the parameter (ΔW/Ao) with a small-the better method is illustrated in the Figure 2, revealing that the overcut smallest value is accomplished at (50 hr) time, (800ºC), and alloy kind (A) (Alloy1).

Table 1. Design of Experiment for the parameters of oxidation process and levels

No.	Alloy symbol	Alloy type	Time (hr)	Temp. ◦C
1	A	1	1	1
2	A	1	2	2
3	A	1	3	3
4	A	1	4	4
5	B	2	1	2
6	B	2	2	1
7	B	2	3	4
8	B	2	4	3
9	C	3	1	3
10	C	3	2	4
11	C	3	3	1
12	C	3	4	2
13	D	4	1	4
14	D	4	2	3
15	D	4	3	2
16	D	4	4	1

In this study, technique of Taguchi design is employed for estimating the optimal weight parameter of gain /area (ΔW/Ao) in the process of oxidation of the α-brass alloys for obtaining the least oxidation layer thickness. Taguchi's orthogonal array experimental design of (L16) was utilized for determining the S/N ratio. The technique of Taguchi was utilized to analyze the influence of every parameter (temperature, time, and type of alloy) upon the generation of oxidation and to foresee the optimal selection for every parameter and analyze the influence of such parameters upon the parameter of (ΔW/Ao). Table 2 lists the input and output results of the oxidation parameters and their levels.

Table 2. Input and output results for Oxidation process parameters

No.	Alloy	Time(hr)	Temp. ºC	$\Delta W/A_o$ (mg/cm2)	SNRA1
1	A	1	500	0.5	6.0206
2	A	10	600	8	18.0618-
3	A	30	700	38	31.5957-
4	A	50	800	95	39.5545-
5	B	1	600	0.5	6.0206
6	B	10	500	4	12.0412-
7	B	30	800	32	30.1030-
8	B	50	700	22	26.8485-
9	C	1	700	1	0.0000
10	C	10	800	28	28.9432-
11	C	30	500	5	13.9794-
12	C	50	600	24	27.6042-
13	D	1	800	1	0.0000
14	D	10	700	8	18.0618-
15	D	30	600	16	24.0824-
16	D	50	500	8	18.0618-

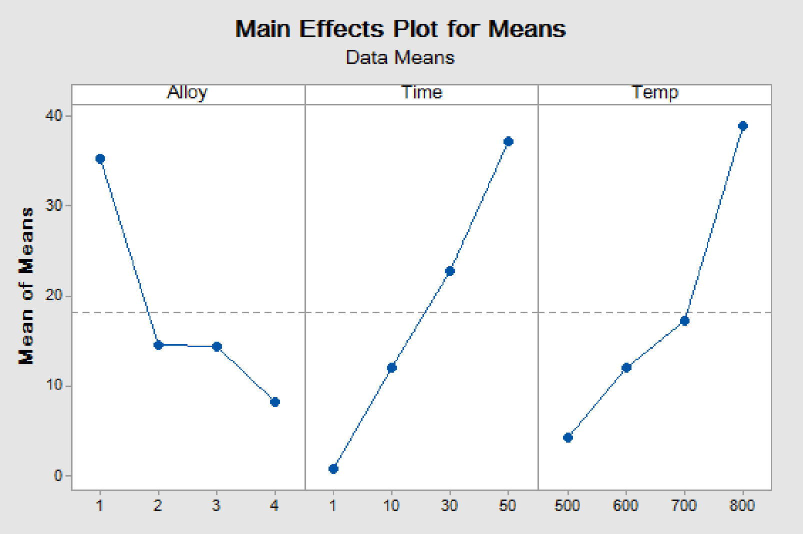

Figure 1. Main effect plot for mean of oxidation process parameters

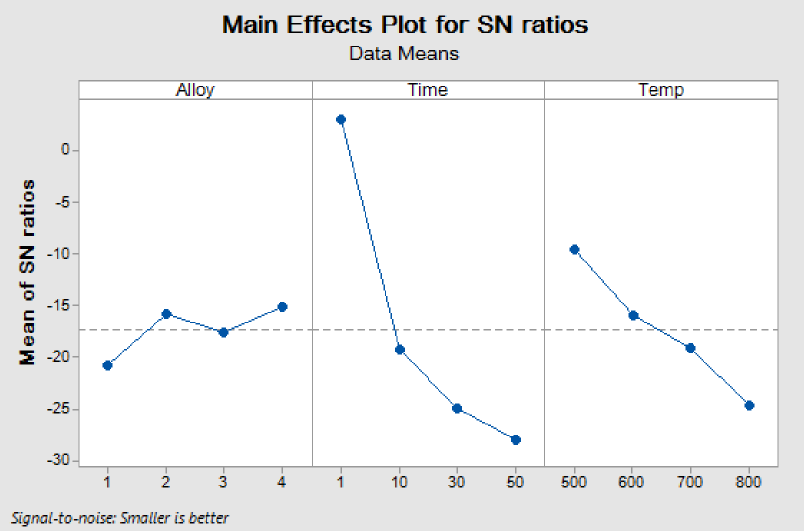

Figure 2. Main effect plot for S/N ratios of oxidation parameters

Analysis of Variance (Anova)

The analysis of weight gain/area (ΔW/Ao) results by Minitab 16 is shown in Table 3. The experimental outcomes were analyzed via the Analysis of Variance (ANOVA) for determining the influence of the oxidation process parameters and the type of alloy containing each one of oxide particles (0.2%TiO_2, 0.2%Y_2O_3 and 0.2%Al_2O_3) on the weight gain/area (ΔW/Ao) as the dependent variable. It was shown that the dominant factor is the oxidation time. Therefore, the most influential factors (parameters) are the the oxidation time and temperature (33.78 and 30.9 %), respectively, while the type of alloy containing the oxide particles had a lower effect (19.94%) on the weight gain/area, as shown in Figure 3. Regression equations (2, 3 ,4 and 5) are for four alloys (A, B,C and D) respectively. Minitab 16 gave imperial equation that denotes the last equation of model depicted in the equation 6. The last equation of model can be utilized with any time and temperature as well as brass alloy type.

Regression equations for four alloys (A, B,C and D) are following:

Alloy (A): $(\Delta W/Ao) = -77.6 + 0.143\ \text{Temperature} + 0.896\ \text{Time}$ (2)

Alloy (B): $(\Delta W/Ao) = -89.8 + 0.143\ \text{Temperature} + 0.896\ \text{Time}$ (3)

Alloy (C): $(\Delta W/Ao) = -101.1 + 0.143\ \text{Temperature} + 0.896\ \text{Time}$ (4)

Alloy(D): $(\Delta W/Ao) = -83.3 + 0.143\ \text{Temperature} + 0.896\ \text{Time}$ (5)

$(\Delta W/Ao) = \text{MEAN1} = -48.4 - 8.15\ \text{Alloy} + 0.706\ \text{Time} + 0.1090\ \text{Temperature}$ (6)

Table 3. Analysis of Variance (ANOVA)

Variance Source	D.O.F	Squares Sum	Variance	P(%)
Alloy	3	1696	565.333	19.94
Time(sec)	3	2871	957	33.78
Temp(C)	3	2627	875.666	30.90
Error ,(e)	7	1307	186.71	15.37
Total	15	8501		100

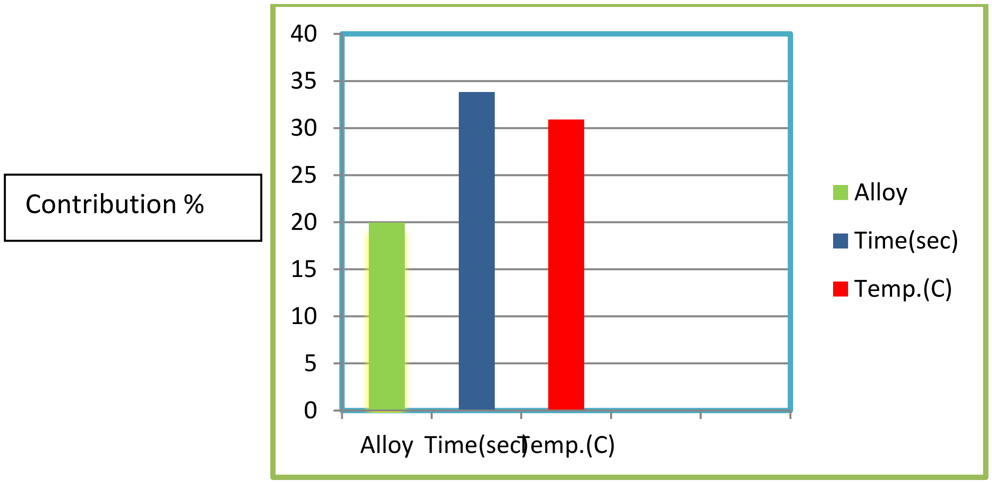

Figure 3. Influential effects based on percentage of distributions.

Effect of Temperature, Time And Alloy Type On (*Δw/Ao) Parameter*

The combination effects of temperature, time and alloy type on the weight gain/area (ΔW/Ao) parameter are represented in Figures 4, 5 and 6 that indicated a 3D contour plot. It was found that the maximum effect on the (ΔW/Ao) or oxidation rate was 50 hr, 800ºC and base alloy (A) (Alloy 1). This is due to increasing the diffusion process of Zn^{+2} ions to the brass surface and reacting with the O_2 gas forming (ZnO) unprotected layer on the surface. This process increases continuously with the time and temperature leading to a greater oxidation rate for all studied alloys in this work. But, the (ΔW/Ao) values of alloys containing oxide particles (0.2%TiO_2, 0.2%Y_2O_3 and 0.2%Al_2O_3) separately were smaller than that of base alloy (A) at the all temperatures (500-800)ºC.

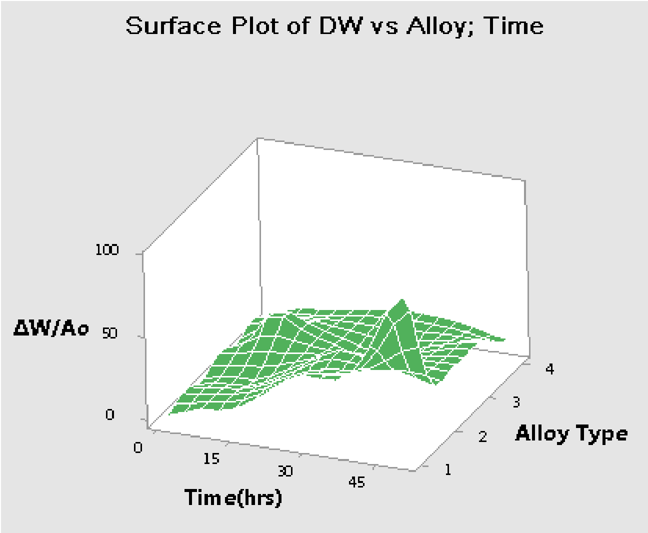

Figure 4. Contour plot of weight gain (ΔW/Ao) vs. time and alloy type

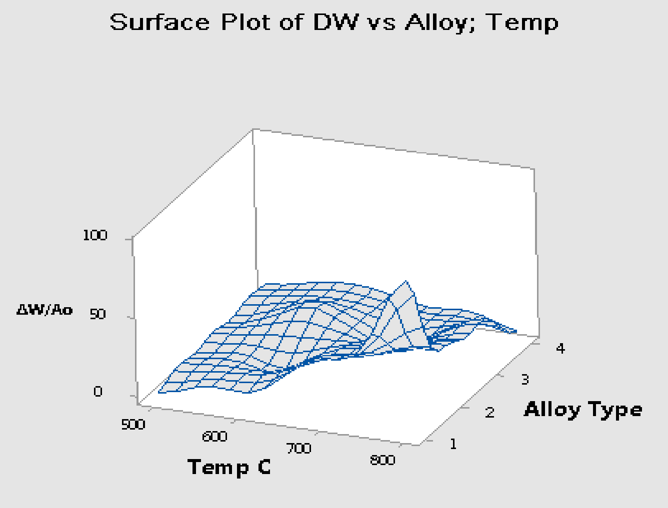

Figure 5. Contour plot of weight gain (ΔW/Ao) vs. temperature and alloy type

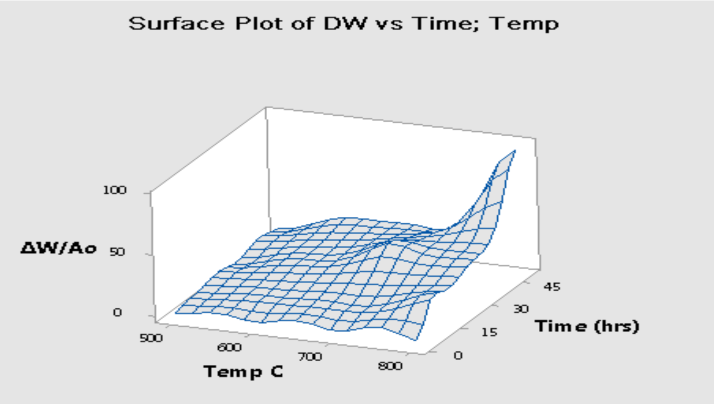

Figure 6. Contour plot of weight gain (ΔW/Ao) vs. temperature and time

Oxidation Behavior of Alloys (A, B, C And D)

From Figure 7, it was revealed that during the measurements of the isothermal oxidation the weight gain raises with the time, because through the oxidation primary period ZnO and CuO are developed upon the specimen (Alloy A) surfaces, also as the oxidation lasts, the layers of oxide will be uninterrupted and separate the (Al_2O_3) from alloy (D). While, TiO_2 and Y_2O_3 oxides are formed in alloys B and C, respectively. Also, change of the forms of the gain/weight area ($\Delta W/Ao$) variation versus the curves of time for the alloy (A) is owing to the increased oxide layers quantity that are shaped upon such specimens surface, as illustrated in Figure 7. The obtained results for the alloy (A) manifested that the kinetics of oxidation of such alloy is governed via the Zn interdiffusion throughout the scale of CuO, and the interface reaction of oxide/gas includes the development of various oxides (CuO, Cu_2O, and ZnO) scales on the brass alloy surface. The kinetics of oxidation for the alloy (A) follows a parabolic law at the measurement of weight gain/ area ($\Delta W/Ao$). Nevertheless, this law of oxidation rate was reached for the oxidized specimens at (500°C) and (800°C) beyond (50 hr) of the time of oxidation.

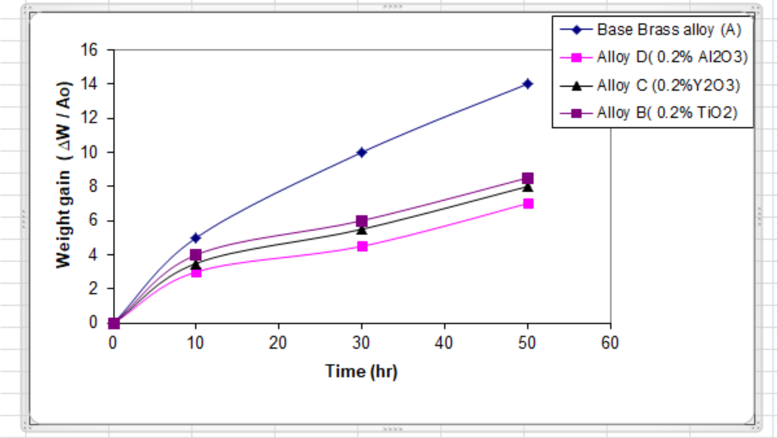

Figure 7. The weight gain /area ($\Delta W/Ao$) versus the time for different alloy types at oxidation temperature of 800ºC

The results comparison for the alloy (A) (70Cu/30Zn) oxidation was obtained to those alloys containing each one of oxide particles of 0.2%TiO_2, 0.2%Y_2O_3 and Al_2O_3 (alloys B, C and D), correspondingly. It was obtained that the oxide particles dispersion within the base alloy matrix enhanced the resistance to oxidation and elucidated that the alloy (C) weight gain/area ($\Delta W/Ao$) was lesser than alloy (A) at the whole temperatures of oxidation. That is owing to the (Y_2O_3) particles role in decreasing the rate of oxidation in base alloy (A) and reducing the rate of (ZnO) scale growth and other layers of oxide, Because the atoms of (Y_2O_3)enhanced the adhesion between the scale and the substrate alloy via double mechanisms [19, 20]. The alloy (D) containing 0.2% Al_2O_3 was the best alloy that had the lowest oxidation rate at all temperatures as compared to other alloys.

CONCLUSIONS

It was found that the oxidation rate or weight gain/surface area ($\Delta W/Ao$) increases continuously with increasing time and temperature for all investigated alloys at (500-800ºC) for 50 hr in dry air.

The addition of small percent of (0.2wt%) oxide particles of (TiO_2, Y_2O_3 and Al_2O_3) separately to the alpha brass alloy improved the oxidation resistance at all oxidation temperatures in various values of weight gain ($\Delta W/Ao$).

It was found that 0.2% Al_2O_3 containing alloy gave the lowest oxidation rate as compared to the base alloy and alloys containing Y_2O_3 and TiO_2 at all temperatures.

From the ANOVA analysis, it was obtained that the contribution percentage was 33.78% for oxidation time followed by oxidation temperature 30.9% and alloy type (modified brass) 19.94%.

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