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Obtaining AlMg/AlN Composites and Using Matrix Modeling Based on Taguchi Method

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Abstract
The aim of the experimental research conducted as part of this paper was to develop new „in situ" composite materials with aluminum alloy matrices reinforced with aluminum nitride particles, by controlling the process variables to ensure that the resulting composite material exhibits improved properties compared to the matrix alloy. We conducted a study on the „in situ" laboratory-scale production of metal composites with a single type of reinforcement particles (AlN), by controlling a series of technological parameters: melt temperature, magnesium content in the matrix alloy, diameter of the reactive gas injection nozzles, gas flow rate, and bubbling duration. This scientific work aimed to determine the significance of various parameters in the process of producing AlMg/AlN composites. Using the Taguchi method, the degree of significance of the analyzed factors influencing the production of composites meeting specific requirements was studied. Moreover, the mentioned method facilitates the design and implementation of an experimental testing plan, through which the empirical mean values of the key parameters were determined.

Keywords: *„In situ"composite materials, Gas bubbling, Taguchi method.*

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I. INTRODUCTION

In the current economic context, where dependence on oil and natural gas controls every aspect of our lives, a better appreciation of the efficient use of these resources is necessary. The high demand in industry for cost-effective and high-performance metal matrix composite materials involves the development of manufacturing and processing methods that are economically and ecologically efficient. Aluminum matrix composite materials (AlMCMs), which exhibit high mechanical strength and wear resistance properties, represent a valuable resource for such materials. Specifically, particle-reinforced MMCM (metal matrix composite materials) present greatest interest due to their ease of manufacturing, low cost and isotropic properties [1÷4].

It has been found that the properties of AlMCMs can be controlled by the size of the reinforcing particles and their volumetric fraction, as well as by the characteristics of the matrix-reinforcement interface. Particle-reinforced AlMCMs are known for their use in wear-resistant parts (large-sized particles) and structural parts (small-sized particles). However, small-sized particles (less than 10 μm) are considerably more difficult to obtain and to incorporate into metallic matrix using traditional processing methods and are significantly more expensive than larger particles. Casting technologies such as rheocasting, pressure casting, compocasting, along with powder metallurgy, preform infiltration, spraying, and mechanical alloying, represent the main methods for obtaining particle-reinforced AlMCM. Within these technologies, reinforcement particles serve as the raw material for the process and are subsequently added to the matrix to produce the final composite. To achieve high and durable mechanical properties for competitive AlMCMs, it is essential to establish a stable interface between the matrix and the reinforcement. Common issues with traditional methods include the complex and complementary particle surface treatment stages, which further increase the final production costs of the composite.

The newly emerged „in situ" technologies, such as DIMOX (Direct Metal Oxidation), PRIMEX (Pressureless Metal Infiltration), XD (Exothermic Dispersion), SHS (Self-Propagating High-Temperature Synthesis), and RD (Radiation Defects), produce aluminum matrix composites with high volumes of reinforcement particles like SiC, AlN, TiC, TiB_2 , etc. Although XD and SHS methods offer lower processing costs due to reduced energy consumption, they involve multiple and less controllable processes. Composites obtained through these methods exhibit uncertain durability due to interfacial instability, may contain undesirable secondary compounds, and often have high porosity. Other methods, such as DIMOX, PRIMEX, and RD, involve the use of presintered reinforcement particles as part of multi-step processes, significantly increasing the projected production costs.

Among the techniques for synthesizing AlMCMs, Reactive Gas Injection (RGI), in which reinforcement particles are formed „in situ" within the molten alloy through an exothermic gas-liquid reaction, is highly attractive due to its simplicity, cost-efficiency, flexibility, and the uniform distribution of small particles in the final composite. The RGI method involves injecting a reactive gas (such as methane, nitrogen, or ammonia) into the molten alloy to produce a composite material through chemical reactions occurring at the liquid alloy–gas bubble interface. Previous studies have successfully demonstrated the production of Al-Si/SiC, Al/AlN, Al/TiC, and Al-TiN composites using the reactive gas injection method [5].

II. THE TECHNOLOGICAL PROCESS FOR OBTAINING AlMg/AlN COMPOSITES THROUGH REACTIVE GAS INJECTION (RGI)

The experimental procedure for obtaining AlMg/AlN composite materials is based on the „in situ" method and involves bubbling nitrogen into the AlMg melt to form AlN reinforcement particles. For this purpose, a closed-system installation is used, which allows the injection of the reactive gas into the molten metal, protected by an inert atmosphere.

Each experiment involves completing at least five main steps:

- Producing and casting semi-finished Al-Mg alloy products that will serve as the raw material for the „in situ" processing installation;
- Loading the alumina crucible with the previously prepared raw material;
- Electromagnetic induction heating of the working chamber to an operating temperature of 950–1000°C and maintaining this temperature throughout the bubbling process;
- Bubbling nitrogen (the reactive gas) into the molten matrix alloy;
- Cooling the system until the composite material in the crucible reaches room temperature.

As a result of the chemical reaction between the nitrogen in the bubbled gas and the aluminum in the molten alloy, micron-sized and thermodynamically stable AlN particles are formed.

The nitrogen is injected at the bottom of the molten alloy column to ensure the gas bubbles travel the longest possible path through the metal melt. Studies on the kinetics of AlN formation have revealed that the slowest step is the chemisorption of nitrogen at the gas/liquid interface. Consequently, both the process temperature and the purity of the reactive gas introduced into the system significantly influence the progression of the process.

To improve the purity of the bubbled gas, preliminary filtration was performed. Magnesium alloying also plays a significant role in the kinetics of nitride formation processes, as magnesium acts as a surfactant and deoxidizer in these systems.

The technological flow outlining the stages of the process for producing composite materials with Al alloy matrices obtained „in situ" through reactive gas bubbling is shown in Figure 1.

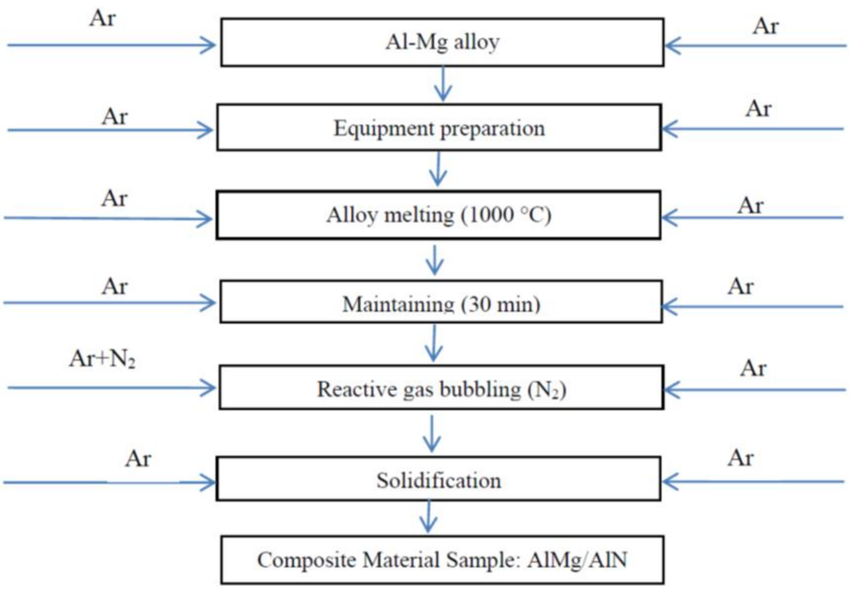

Figure 1: The process flow diagram for in situ production of composites through reactive gas bubbling

III. RESULT AND DISCUSSION

The key parameters influencing the processes within the system of molten and overheated aluminium alloy and gaseous nitrogen are:

- Holding temperature of the alloy column during the entire nitrogen bubbling period;

- Magnesium concentration in the aluminium alloy bubbled with nitrogen;
- Flow rate of the reactive gas (nitrogen) injected at the base of the aluminium alloy column;
- Bubbling time and nitrogen injection into the metallic melt;
- Gas injection nozzle sizes of 0.8 mm, 1.0 mm, and 1.2 mm;
- Liquid alloy column height.

For the present work, a total of 16 batches of composite material were developed. For each batch, the technological parameters were varied. Table 1 illustrates the technological parameters used for the 16 experiments.

Table 1: Experimental Parameters

Batch No.	Composite Material Type	Temperature, ºC	Percent of Mg, %gr.	Gas Flow Rate, l/min.	Bubbling Time, min.	Blowing Hole Diameter, mm.	Melt Column Height, mm.
1.	AlMg15/AlN	1000	15	0,6	360	1,2	375
2.	AlMg10/AlN	1000	10	0,6	360	1,2	375
3.	AlMg5/AlN	1000	5	0,6	360	1,2	375
4.	AlMg15/AlN	950	15	0,4	180	0,8	375
5.	AlMg10/AlN	950	10	0,4	180	0,8	375
6.	AlMg5/AlN	950	5	0,4	180	0,8	375
7.	AlMg15/AlN	1000	15	0,4	240	0,8	375
8.	AlMg10/AlN	1000	10	0,4	240	0,8	375
9.	AlMg5/AlN	1000	5	0,4	240	0,8	375
10.	AlMg15/AlN	1000	15	0,4	240	1,2	375
11.	AlMg10/AlN	1000	10	0,5	300	1,0	375
12.	AlMg5/AlN	1000	5	0,5	300	1,0	375
13.	AlMg15/AlN	1000	15	0,5	300	1,0	375
14.	AlMg10/AlN	1000	10	0,5	300	1,2	375
15.	AlMg5/AlN	1000	5	0,5	300	1,2	375
16.	AlMg10/AlN	1000	10	0,5	300	1,2	375

IV. MATRIX MODELING OF EXPERIMENTAL RESULTS FOR AlMg/AlN COMPOSITES OBTAINED „IN SITU" THROUGH REACTIVE GAS BUBBLING

The general experimentation and planning methodology used is based on Taguchi. The proposed method seeks to meet certain criteria, such as ease of acquisition, minimization of the number of trials, and therefore the cost of experimentation, while providing the best possible accuracy [6, 7].

Taguchi developed an original method that, starting from a few standard tables, allows for the easy resolution of most industrial problems related to experimental design. While techniques for orthogonal experimental designs are publicly available, Taguchi's originality lies in the implementation strategies, offering a subset of standard arrangements that are sufficient for current practice.

The general methodology approached by the Taguchi method is presented in Figure 2.

The initiation of parameter determination highlights three original aspects:

- reducing effects by leaving unchanged the causes that are impossible or too costly to reduce;
- the main quality criterion of a process is the relative dispersion of its performance;

Taguchi developed linear charts that provide a graphical representation of assigning factors to the columns of an orthogonal arrangement. These charts simplify the implementation of these orthogonal arrangements.

The acquisition of new knowledge for improving the quality of products and processes is based on a progressive process, frequently relying on experimentation.

Experimental designs constitute a method for optimizing the process of acquiring relevant knowledge, and Taguchi's intervention clearly simplifies the procedure for establishing a fractional plan. The Taguchi procedure was designed to improve the performance of a process influenced by numerous factors [8].

The advantages of the Taguchi method are:

- a strict plan for conducting experiments is implemented;
- a considerable reduction in the number of trials compared to traditional techniques;
- the study can include a large number of factors while establishing interactions between factors;
- results are obtained with maximum precision and interpreted without errors;
- a mathematical model of the studied system can be obtained.

The model proposed by Viger and Sisson [6], which is easy to study, presents a matrix model of a system composed of “I” factors: F_1 , F_2 , ... F_i, with each factor having “n_i” levels (equation 1).

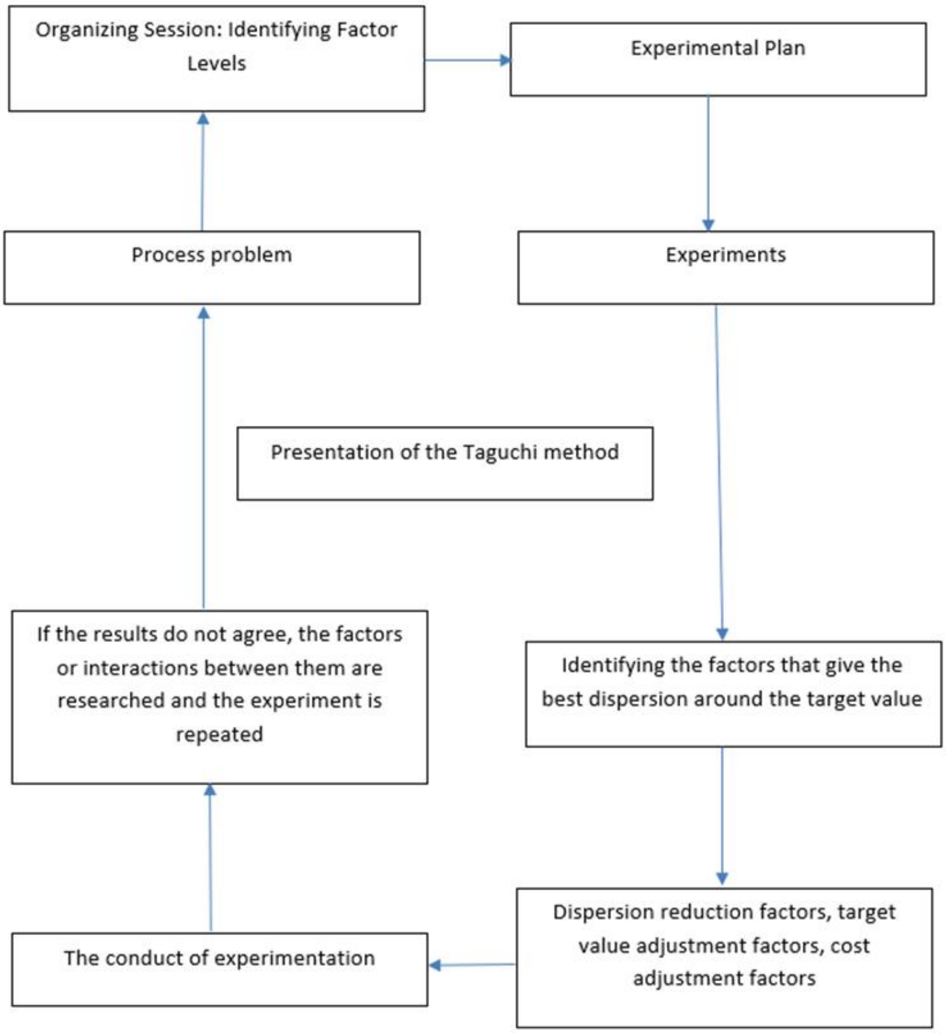

Figure 2: General Methodology Approached by Taguchi [8]

$$Z_t = M + \left\lfloor E_{F_1^1} E_{F_1^2} \dots E_{F_1^{n_1}} \right\rfloor \cdot [F_1] + \left\lfloor E_{F_2^1} E_{F_2^2} \dots E_{F_2^{n_2}} \right\rfloor \cdot [F_2] + \cdots$$
$$+ \left\lfloor E_{F_i^1} E_{F_i^2} \dots E_{F_i^{n_i}} \right\rfloor \cdot [F_1] +.^t [F_1] \begin{vmatrix} I_{F_1^1 F_2^1} & I_{F_1^1 F_2^2} \dots I_{F_1^1 F_2^{n_2}} \\ I_{F_1^2 F_2^1} & I_{F_1^2 F_2^2} \dots I_{F_1^2 F_2^{n_2}} \\ \dots & \dots \quad \dots \\ I_{F_1^{n_1} F_2^1} & I_{F_1^{n_1} F_2^2} \dots \quad I_{F_1^{n_1} F_2^{n_2}} \end{vmatrix}$$
$$\cdot [F_2] + \dots +.^t [F_1] \begin{vmatrix} I_{F_1^1 F_3^1} & I_{F_1^1 F_3^2} \dots I_{F_1^1 F_3^{n_3}} \\ I_{F_1^2 F_3^1} & I_{F_1^2 F_3^2} \dots I_{F_1^2 F_i^{n_3}} \\ \dots & \dots \quad \dots \\ I_{F_1^{n_1} F_3^1} & I_{F_1^{n_1} F_3^2} \dots \quad I_{F_1^{n_1} F_i^{n_3}} \end{vmatrix} \cdot [F_3] + \cdots$$
$$+.^t [F_1] \begin{vmatrix} I_{F_1^1 F_i^1} & I_{F_1^1 F_i^2} \dots I_{F_1^1 F_i^{n_i}} \\ I_{F_1^2 F_i^1} & I_{F_1^2 F_i^2} \dots I_{F_1^2 F_i^{n_i}} \\ \dots & \dots \quad \dots \\ I_{F_1^{n_1} F_i^1} & I_{F_1^{n_1} F_i^2} \dots \quad I_{F_1^{n_1} F_i^{n_i}} \end{vmatrix} \cdot [F_i] + \cdots$$
$$+.^t [F_{i-1}] \begin{vmatrix} I_{F_{i-1}^1 F_i^1} & I_{F_{i-1}^1 F_i^2} \dots I_{F_{i-1}^1 F_i^{n_i}} \\ I_{F_{i-1}^2 F_i^1} & I_{F_{i-1}^2 F_i^2} \dots I_{F_{i-1}^2 F_i^{n_i}} \\ \dots & \dots \quad \dots \\ I_{F_{i-1}^{n_{i-1}} F_i^1} & I_{F_{i-1}^{n_{i-1}} F_i^2} \dots \quad I_{F_{i-1}^{n_i} F_i^{n_i}} \end{vmatrix} \cdot [F_2] \quad (1)$$

where:

Z_t is the theoretical response of the system;

M is the overall mean of the responses, calculated as the ratio between the sum of the response values and the number of experiments conducted;

[F_1] is the vector indicating the level of factor F_i, represented as a column matrix with zero elements except for one, which is equal to 1, and it corresponds to the row „i" corresponding to the level of the factor being considered;

$E_{F_i^j}$ is the average effect of the system's response for factor F_i at level j, and is calculated by subtracting the overall mean M from the system's response mean.

$\left\lfloor I_{F_i^j F_k^t} \right\rfloor$ - the interactions between factors F_i and F_k are calculated by subtracting the overall mean from the system's response mean when factor F_i is at level j and factor F_k is at level t, overall average (M) - $E_{F_i^j}$.

It is very difficult to represent a model that integrates all the factors of a complete system, so it is necessary to resort to the disaggregation of specific factors.

Matrix modelling will be applied using Taguchi's method, and the model will be written according to Viger and Sisson. The goal will be to determine the coefficients of a model of the form (2):

$$Z_t = M + C + t_b + T_{bub} + Q_{gi} + D_i + H + C \cdot t_b + C \cdot T_{bub} + C \cdot Q_{gi} + C \cdot D_i + t_b \cdot T_{bub} \quad (2)$$

where:

M is the general mean;
C is the concentration of Mg, [%];
t_b is the bubbling time [min];
T_{bub} is the bubbling temperature [°C];
Q_{gi} is the gas flow rate [l/min];
D_i is the diameter of the blowing holes [mm];
H is the melt column height [mm].

The number of degrees of freedom for the model is determined by considering that each input parameter is at two levels. Therefore, for the individual parameters, there are 6 (2-1) = 6 degrees of freedom, and for the interactions between the parameters, there are 6 (2-1)(2-1) = 6 degrees of freedom.
The total number of degrees of freedom for the model is given by the following relationship:

$$N_{n,m} = n_g l_n \cdot n_g l_m = (n_i v_n - 1)(n_i v_m - 1) \quad (3)$$

Where:
$n_g l_n$ and $n_g l_m$ represent the number of degrees of freedom for factors n and m;
$n_i v_n$ and $n_i v_m$ represent the number of levels for factors n and m.

Therefore, for the presented model, the number of degrees of freedom is the sum of the degrees of freedom for the effects of the input factors and their interactions, with an additional degree of freedom for the effect of the mean M. Thus, the total degrees of freedom is: 1+6+6=13 degrees of freedom.

Taguchi divides the input factors as shown in Table 2.

Table 2: Groups of Input Factors According to Taguchi [6]

Group I	Group II	Group III	Group IV
Very difficult to modify factors	Difficult to modify factors	Easily modifiable factors	Very easily modifiable factors

For the model presented above, the grouping is as follows (Table 3):

Table 3: Groups of Input Factors for the Studied Model

Group I	Group II	Group III	Group IV
-	D_i; H	T_{bub}; Q_{gi}	t_b, C

Building a fractional plan at the variation levels of the input parameters (Table 4) is not a simple task.

Table 4: Levels of Variation of Input Parameters

Input Parameter Levels	C [%]	T_{bub} [°C]	t_b [min]	Q_{gi} [l/min]	D_i [mm]	H [mm]
Level 1	5	950	180	0,4	0,8	290
Level 2	15	1000	360	0,6	1,2	375

For performing the fractionalization of the experiment, several conditions must be verified. One indispensable condition for calculating the effects of a factor independently of other factors is the condition of orthogonality. Two disjoint actions (which do not share common factors) are orthogonal if, at each level of one, all levels of the other are associated the same number of times in the experimental design. An experimental design is orthogonal with respect to a model if all the disjoint actions of the model are orthogonal in the experimental design. To verify the orthogonality condition, Table 4 was created. After analyzing it, it was concluded that the smallest orthogonal program that can be designed is one that involves 8 experimental trials.

A second condition is to verify the number of degrees of freedom. The number of degrees of freedom of a model indicates the number of values that need to be calculated in order to determine the complete set of model coefficients. It is necessary to conduct at least as many experiments as the number of degrees of freedom in the model. As mentioned earlier, there are 13 degrees of freedom for the model, so the experimental design must include at least 13 trials to properly estimate the effects and interactions of the factors. This ensures that the model's parameters can be independently estimated without any loss of information.

Table 5. Orthogonality Condition

C 2	*										
Q_{gi} 2	2^2	*									
D_i 2	2^2	2^2	*								
t_b 2	2^2	2^2	2^2	*							
T_{top} 2	2^2	2^2	2^2	2^2	*						
H 2	2^2	2^2	2^2	2^2	2^2	*					
CH 2^2	*	*	2^3	2^3	2^2	2^3	*				
CT_{top} 2^2	*	2^3	2^3	2^3	2^3	2^3	2^3	*			
CQ_{gi} 2^2	*	2^3	2^3	2^3	*	2^3	2^3	2^3	*		
CD_i 2^2	*	2^3	2^3	2^3	2^3	*	2^3	2^3	2^3	*	
t_bT_{top} 2^2	2^3	2^3	2^3	*	*	2^3	2^3	2^3	2^3	2^3	*
	2 C	2 Q_{gi}	2 D_i	2 t_b	2 T_{top}	2 H	2^2 CH	2^2 CT_{top}	2^2 CQ_{gi}	2^2 CD_i	2^2 t_bT_{top}

Next, the model's graph is created, shown in Figure 3, which results in the assignment of columns to the independent factors (Table 6).

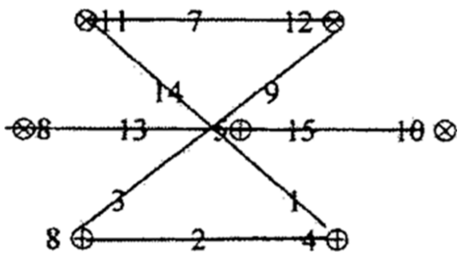

Figure 3: Experimental Graphical Model, Type 2 [3, 9]

Table 6. Assignment of Columns to Independent Factors

Number of Trials	C [%]	Q_{gi} [l/min]	D_i [mm]	t_b [min]	T_{barb} [°C]	H [mm]
1	1	1	1	1	1	1
2	1	1	2	2	2	2
3	2	2	1	1	2	1
4	2	2	2	2	1	2
5	1	1	2	2	1	1
6	1	1	1	1	2	2
7	2	2	2	2	2	1
8	2	2	1	1	1	2
9	2	1	1	2	1	1
10	2	1	2	1	2	2
11	1	2	1	2	2	1
12	1	2	2	1	1	2
13	2	1	2	1	1	1
14	2	1	1	2	2	1
15	1	2	2	1	1	2
16	1	2	1	2	1	2

Table 7. Output Parameters Resulting from Experiments

Nr.	Type of Composite	Particle Size of AlN D_p [μm]	Quantity of Nitride C_{AlN} [% gravimetrice]
1.	AlMg15/AlN	19,5	14,85
2.	AlMg10/AlN	13	11,55
3.	AlMg5/AlN	3,5-4	6,23
4.	AlMg15/AlN	18,5	14,67
5.	AlMg10/AlN	11,5	11,47
6.	AlMg5/AlN	4	6,03
7.	AlMg15/AlN	19	14,54
8.	AlMg10/AlN	12	11,08
9.	AlMg5/AlN	4,5	5,90
10.	AlMg15/AlN	17,5	14,24
11.	AlMg10/AlN	10,5	10,98
12.	AlMg5/AlN	4	5,78
13.	AlMg15/AlN	18	14,15
14.	AlMg10/AlN	11	10,56
15.	AlMg5/AlN	3	5,69
16.	AlMg10/AlN	9	10,79

V. CONCLUSION

Aluminum, magnesium and their alloys do not dissolve nitrogen, but at temperatures much higher than those corresponding to the elaboration, they form nitrides. At normal processing temperatures of 700-750ºC, nitrogen is used for degassing by bubbling.

The „in situ" formation of reinforcement particles in the liquid metal matrix is a promising technique for manufacturing composites from both technical and economic considerations.

For the model presented in the paper, the number of degrees of freedom is the sum of the degrees of freedom of the effects of the input factors and the interactions between them, to which is added a degree of freedom for the effect of the mean M, so 13 degrees of freedom.

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