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THEORETICAL CALCULATION OF THE DENDRITE NON-HOMOGENEITY IN MAGNESIUM-ZINC ALLOYS

RYSZARD CIACH

PHYSICAL METALLURGY DIVISION

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THEORETICAL CALCULATION OF THE DENDRITE NON-HOMOGENEITY IN MAGNESIUM-ZINC ALLOYS

by

Ryszard Ciach*

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ABSTRACT

The distribution of zinc in magnesium-zinc solid solution, and the maximum amount of Mg_7Zn_3 compound due to the non-equilibrium freezing of the Mg-6 wt% Zn, were calculated.

The calculation was carried out according to Krupkowski's equations which assume complete diffusion in the liquid state and no diffusion in the solid state and take into account the changes of the distribution coefficient during the solidification.

The results of calculation indicate that:

- there is a large region inside the grains containing approximately 0.74 wt% Zn, which increases significantly only near the Mg-Mg_7Zn_3 eutectic.
- out of the original 6 wt% Zn in the alloy, 4.077 wt% Zn may form Mg_7Zn_3 compound and only 1.923 wt% Zn will be in solid solution.

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Direction des mines

Rapport de recherche R 203

CALCUL THÉORIQUE DE L'HÉTEROGÉNÉITÉ DENDRITIQUE DANS LES ALLIAGES DE MAGNÉSIUM-ZINC

par

Ryszard Ciach*

RÉSUMÉ

L'auteur a calculé la répartition du zinc dans une solution solide de magnésium ainsi que la quantité maximale de Mg_7Zn_3 causée par le déséquilibre qui se produit au cours du refroidissement du mélange Mg-6%Zn (poids).

Le calcul a été effectué selon les équations de Krupkowski qui supposent la diffusion complète à l'état liquide et l'absence de diffusion à l'état solide et tiennent compte des changements du coefficient de répartition au cours de la solidification.

Les résultats du calcul indiquent:

- qu'une zone importante à l'intérieur des grains contient environ 0.74% de Zn (en poids), pourcentage qui ne s'accroît de façon significative que près de l'eutectique $Mg-Mg_7Zn_3$;
- que du pourcentage original de 6% de Zn (en poids) de l'alliage, 4.077% (en poids) de Zn peut contribuer à former le composé Mg_7Zn_3 et seulement 1.923% (en poids) de Zn sera en solution solide.

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INTRODUCTION

The structure of the Mg-Zn casting alloys usually shows unequal distribution of the zinc within the grains of magnesium-zinc solid solution and, also, the presence of a non-equilibrium compound Mg_7Zn_3. A previous investigation at the Physical Metallurgy Division of the Mines Branch by Lagowski [1] has shown the presence of this compound in cast Mg-Zn alloys with as little as 2 wt% Zn. This compound is harmful because it ties up the zinc, which otherwise would be in solid solution; therefore, a knowledge of its occurrence is desirable.

The dissolution of this compound in Mg-solid solution is one of the important factors to be considered in determining the suitable methods and parameters of heat treatment to achieve optimum properties in these alloys. As an example, the high mechanical properties were obtained by Meier [2, 3, 4, 5] and Lagowski [3, 5] in Mg-Zn-Zr and Mg-Zn-Ag-Zr alloys by a special high-temperature solution heat treatment.

The present report will attempt to determine the maximum degree of the segregation of the zinc within the grains, as well as the maximum amount of the Mg_7Zn_3 phase formed as the result of the most unfavourable freezing conditions.

BASIS OF CALCULATIONS

The calculation has been carried out according to Krupkowski's equations [6, 7, 8], which make the following assumptions:

- Complete diffusion in the liquid.
- No diffusion in the solid.
- A sufficiently high solidification rate to ensure that the composition of layers forming on the surface of dendrites corresponds to the solidus line.

The method also takes into account the changes of the distribution coefficient during the freezing process.

All these conditions are considered necessary to achieve maximum degree of segregation.

In the calculation, the data from the Mg-Zn equilibrium diagram according to Adenstedt and Burns [9] were used. The eutectic point and the maximum solid-solubility were modified according to the results of Clark and Rhines [10], Meier [11], and Park and Wyman [12]. The Mg-Mg_7Zn_3 part of this diagram is presented in Figure 1, and all calculations are based on this quasi-binary system.

The calculation is divided into two parts:

I. That which deals with the solidification data within the ranges into which the Mg-Mg_7Zn_3 system has been divided.

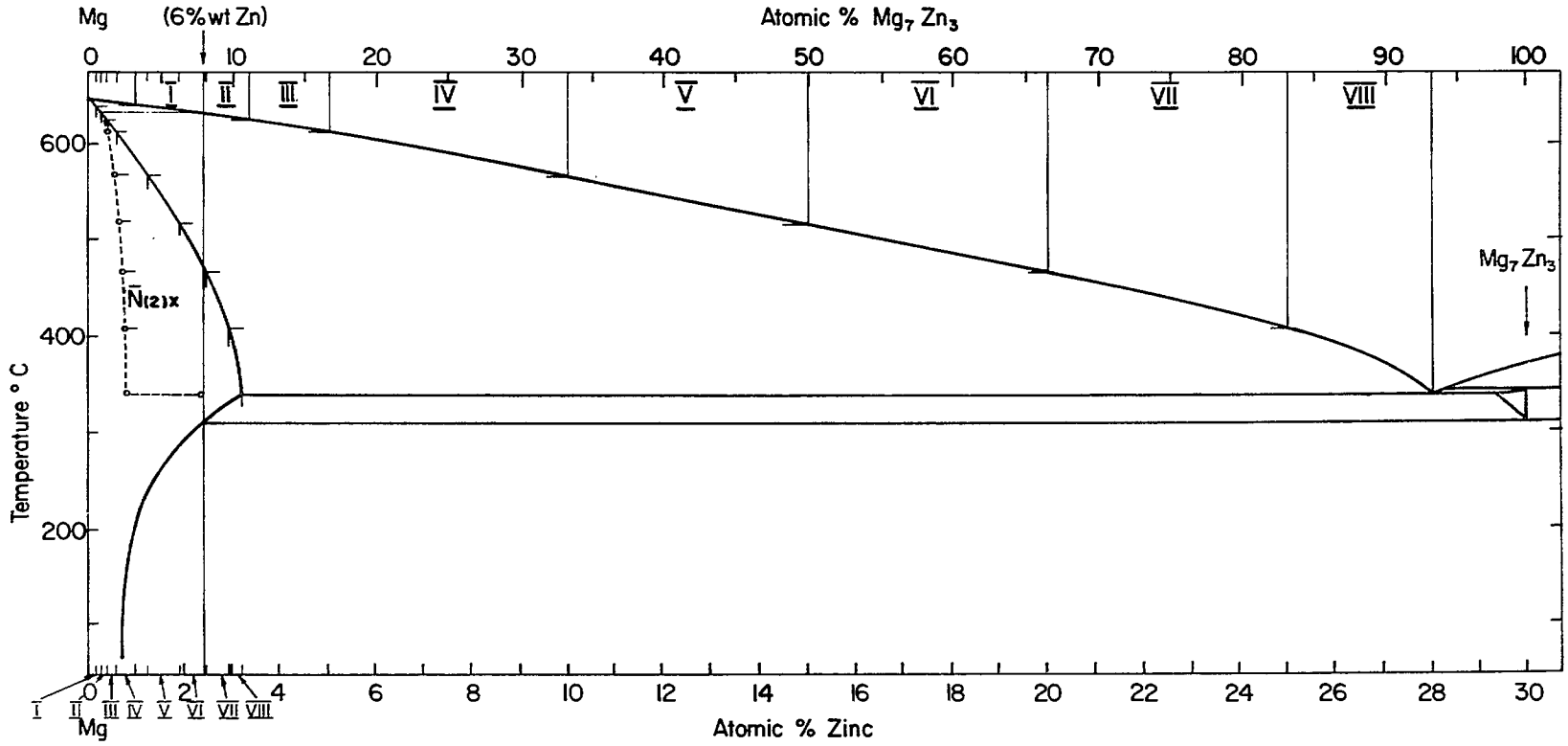

Figure 1. System $Mg-Mg_7Zn_3$ and changes of over-all composition of dendrites ($\bar{N}_{(2)x}$) in Mg- 6 wt% Zn alloy.

II. That which deals with the solidification process for a particular alloy, Mg-6 wt% Zn, using the data from the first part. Similarly, the same data from Part I may be used for any other Mg-Zn alloy.

Part I - Solidification Data for Ranges I-VIII in Mg-Mg_7Zn_3 System

The Mg-Mg_7Zn_3 system has been divided into eight ranges, designated I to VIII, and the limits of these ranges are nine alloys designated 1 to 9, compositions of which are presented in Table 1 and shown in Figure 1.

The compositions and quantities throughout the calculations are given in mole fractions, assuming that the amount of alloy is one mole. However, as compositions and quantities taken from the Mg-Mg_7Zn_3 equilibrium diagram can be expressed in terms of either Mg-(1) or Mg_7Zn_3-(2), the sum of both such complementary terms is one.

The following designations are used throughout the paper, brackets round the subscripts indicating the solid state and the absence of them the liquid state.

N_1, N_2 - the initial composition of the liquid in terms of Mg, i.e., N_1, or Mg_7Zn_3, i.e., N_2, before solidification starts.

n_1, n_2 - the quantities of the initial liquid in terms of Mg or Mg_7Zn_3.

TABLE 1

The Changes in the Distribution Coefficient (K_{calc}), the Amounts of Dendrites (X), and the Over-all Quantity of Liquid Solidifying as the Eutectic (X_E), in the Mg-Mg_7Zn_3 Quasi-binary System

(All values given in mole fractions)

Alloy Number	N_{2x}	$N_{(2)x}$	K_{calc} and K_{theor}	Range Number	Amount of Dendrites-X	The Constant Values Within the Ranges			X_E
						a	b	c	
1	0.0333	0.0040	0.1212						
				I	0.63317	30.03	-24.259	2.1634	0.02201
2	0.0800	0.0092	0.1150						
				II	0.30607	12.5	-10.255	0.8075	0.06001
3	0.1103	0.0139	0.1260						
				III	0.37645	9.066	- 7.0751	0.8484	0.08648
4	0.1666	0.0206	0.1236						
				IV	0.54875	6.00	- 4.8007	0.4595	0.13870
5	0.3333	0.0416	0.1248						
				V	0.37164	3.00	- 2.3663	0.2594	0.30738
6	0.5000	0.0634	0.1268						
				VI	0.28026	2.00	- 1.5290	0.2173	0.48918
7	0.6666	0.0810	0.1215						
				VII	0.22425	1.5	- 1.1675	0.1503	0.67966
8	0.8333	0.1000	0.1200						
				VIII	0.12385	1.2	- 0.8750	0.1809	0.87614
9	0.9366	0.1053	0.1124						

$N_{(1)x}$, $N_{(2)x}$ - the composition of the solid forming on the surface of the dendrite at a certain solidification stage.

$n_{(1)x}$, $n_{(2)x}$ - the over-all quantities of the components solidified at a certain solidification stage.

N_{1x}, N_{2x} and n_{1x}, n_{2x} - the compositions and quantities of the components in the liquid state at a certain solidification stage which are in equilibrium with the respective $N_{(1)x}$, $N_{(2)x}$ and $n_{(1)x}$, $n_{(2)x}$.

X - the quantities of the solidifying dendrites at a certain solidification stage.

$\bar{N}_{(1)x}$, $\bar{N}_{(2)x}$ - the over-all composition of the components in the dendrites.

The process of solidification within the first range refers to the solidification of the alloy 1 (0.033 mole fraction of Mg_7Zn_3) within the limits of the change in the liquid composition from 0.033 (N_{21}) to 0.08 mole fraction of Mg_7Zn_3 (N_{22}). The fraction solidified in the first range (X_I) is calculated according to the equation:

$$-\frac{\left[A_1(1-X_I) - A_2\right]}{X_I}\ln(1-X_I) = A_1 - A_2 + \frac{1}{N_{21}} - \frac{1}{N_{22}} \qquad \ldots (1)$$

where

$$A_1 = \frac{N_{(2)1} - N_{21}}{N_{21}^2}\,;\quad A_2 = \frac{N_{(2)2} - N_{22}}{N_{22}^2}\,.$$

The right side of Equation 1 can be obtained on the basis of the data from Table 1. The further solution of the equation may be carried out by a graphic or a successive approximation method.

Thus calculated, the values of the X_I to X_{VIII} define the quantity of dendrites forming within the ranges I to VIII and are presented in Table 1.

The second Krupkowski equation [6, 7] defines the changes in the compositions of the liquid according to the amounts of the dendrites:

$$N_{2x} = 1/\left[a + b\,X + c\ln (1 - X) \right] \qquad \ldots \quad (2)$$

where, for example, within the first range,

$$a = \frac{1}{N_{21}} ,$$

$$b = \frac{A_1 - A_2}{X} ,$$

$$c = \frac{A_1 (1 - X) - A_1}{X} .$$

Values a, b, c are constant within each solidification range [8].

Using Equation 2, the composition of the liquid within each range was calculated for arbitrarily chosen values of X up to the maximum value calculated from Equation 1.

The distribution coefficient (K) within the ranges is defined by the following next equation:

$$K_{calc} = 1 + (A - bX)\, N_{2x}\,, \qquad \ldots (3)$$

which should be in agreement with the distribution coefficient obtained from the equilibrium diagram according to the formula

$$K_{theor} = \frac{N_{(2)x}}{N_{2x}} \,. \qquad \ldots (4)$$

The agreement of the K values obtained from Equations 3 and 4 for the limit values of the ranges is a test of correctness for both the X and N_{2x}.

An example of the detailed calculation of range II is presented in Appendix A, and the values obtained for all eight ranges are given in Table 1.

The knowledge of the X for ranges I to VIII lets us define the amount of liquid solidified as a eutectic for the several alloys limiting the solidification ranges, according to Equation 1:

$$X_E = (1 - X_n)\ (1 - X_n + 1) \ldots \qquad (1 - X_{VIII}) \qquad \ldots (5)$$

where X_E is the fraction solidified as eutectic for the alloy that is the upper limit of the range, denoted as n.

The calculation of the amount of liquid solidified as the eutectic for Mg- 6 wt% Zn is presented in Appendix A, and the results of the calculations for the remaining alloys 1 to 9 are shown in the last column of Table 1.

Part II - Solidification Process of Mg-6 wt% Zn Alloy

The detailed calculation of the solidification process for Mg-6 wt% Zn alloy was carried out using previous results and Krupkowski's equations from (6) to (11), which define $N_{(1)x}$, $N_{(2)x}$, $n_{(1)x}$, N_{1x}, N_{2x}, n_{1x}, n_{2x}, $\bar{N}_{(1)x}$ and $\bar{N}_{(2)x}$ values as a function of the fraction solidified (X).

The calculation uses the previous ranges but starts from range II, the upper limit of which corresponds to the Mg-6 wt% Zn alloy. The first values calculated are the quantities of the fraction solidified for the respective changes in the composition of the liquid from 0.08 to 0.9366 mole fraction of Mg_7Zn_3 (eutectic point). The calculation of this increasing fraction solidified is partially based on the previous data in Table 1, as follows:

X_1 - the fraction solidified in the present first range 1 (previous range II) is taken from Table 1. It corresponds to the amount of dendrites 0.30607 mole fraction of Mg_7Zn_3 and thus:

$$X_1 = X_{II}$$

X_2 - the overall fraction solidified within the present ranges 1 and 2 (previous ranges II and III) is calculated, using the following equation:

$$X_2 = 1 - (1 - X_1)\ (1 - X_{III}) \qquad \ldots (6)$$

where X_{III} is the fraction solidified in the previous range III

X_3 - the over-all fraction solidified within the ranges 1, 2 and 3 is calculated as follows:

$$X_3 = 1 - (1 - X_2)\ (1 - X_{IV}) \quad \ldots (6a)$$

and so on up to range 7, where

$$X_7 = 1 - (1 - X_6)\ (1 - X_{VIII}) \quad \ldots (6b)$$

The next values are a function of the X, and are calculated as follows;

$$n_{2x} = N_{2x}\ (1 - X) \quad \ldots (7)$$

$$n_{(2)x} = N_{21} - n_{2x} \quad \ldots (8)$$

where N_{21} is the initial composition (Mg- 6 wt% Zn) ;

$$n_{1x} = 1 - X - n_{2x} \quad \ldots (9)$$

$$n_{(1)x} = X - n_{(2)x} \quad \ldots (10)$$

$$\overline{N}_{(2)x} = \frac{n_{(2)x}}{X} \quad \ldots (11)$$

The detailed calculation of the X, as well as the next values, is shown in Appendix B. The results of the calculation are given in Table 2 and some of them are plotted in Figures 1 and 2.

TABLE 2

Solidification Data for the Mg-6 wt% Zn Alloy up to Eutectic Point
(0.9366 Mole Fraction of Mg_7Zn_3)

N_{2x}	$N_{(2)x}$	X	n_{2x}	$n_{(2)x}$	n_{1x}	$n_{(1)x}$	$\bar{N}_{(2)x}$
0.0800	0.0092	0	0.08000	0	0.92000	0	
0.1103	0.0139	0.30607	0.07654	0.00346	0.68626	0.30262	0.01277
0.1666	0.0206	0.56730	0.07208	0.007913	0.42480	0.55939	0.01395
0.3333	0.0416	0.80474	0.06508	0.01492	0.13017	0.78982	0.01854
0.5000	0.0634	0.87731	0.06134	0.01865	0.06134	0.85866	0.02126
0.6666	0.8100	0.91170	0.05886	0.02114	0.02944	0.89056	0.02318
0.8333	0.1000	0.93150	0.05708	0.02292	0.01142	0.90858	0.02460
0.9366	0.1053	0.94000	0.0562	0.0238	0.00380	0.91620	0.02530

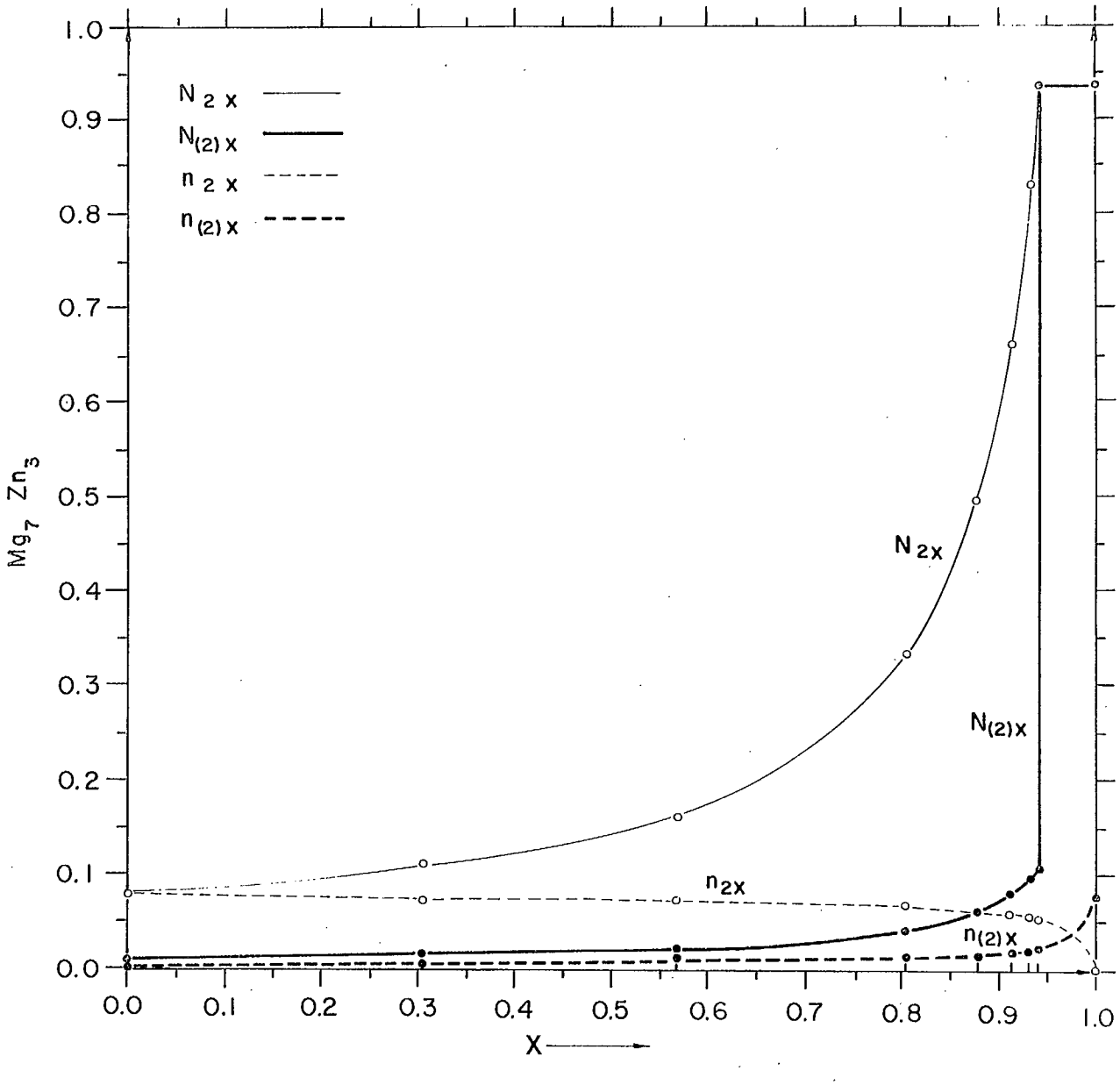

Figure 2. The N_{2x}, $N_{(2)x}$, n_{2x} and $n_{(2)x}$ values versus fraction solidified X, for Mg-6 wt% Zn alloy.

The $N_{(2)x}$ line in Figure 2 shows the change in composition across the dendrites and the existence inside them of the wide region containing a small nearly constant amount of zinc, which rises significantly only afterward near the eutectic.

The maximum amount of the Mg_7Zn_3 compound formed due to the non-equilibrium freezing of the 0.08 mole fraction Mg_7Zn_3 alloy is, according to Table 2, 0.0562 mole fraction Mg_7Zn_3. This means that approximately 4.077 wt% Zn is tied up as Mg_7Zn_3 compound and only 1.923 wt% Zn is in solution.

The possibility of so significant an amount of zinc being tied up as Mg_7Zn_3 compound indicates the importance of finding methods to dissolve the compound and obtain homogeneous solid solution.

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APPENDIX A

Calculation of Solidification Data for Range II

The calculation of the solidification data for range II, for the alloy containing 0.08 mole fraction of Mg_7Zn_3 (Mg-6 wt % Zn) within the limits of the change in the liquid composition from 0.08 to 0.1103 Mg_7Zn_3, is as follows:

The fraction solidified for this range (X_{II}) is calculated from Equation 1:

$$-\frac{\left[A_2(1 - X_{II}) - A_3\right]}{X_{II}} \ln (1 - X_{II}) = A_2 - A_3 + \frac{1}{N_{22}} - \frac{1}{N_{23}} ; \qquad (1)$$

where, using the data from Table 1,

$$A_2 = \frac{N_{(2)2} - N_{22}}{N_{22}^2} = \frac{0.0092 - 0.08}{0.08^2} = -11.0625 ;$$

$$A_3 = \frac{N_{(2)3} - N_{23}}{N_{23}^2} = \frac{0.0139 - 0.1103}{0.1103^2} = -7.92366 ;$$

thus

$$-\frac{\left[-11.0625 (1 - X_{II}) + 7.92366\right]}{X_{II}} \ln (1 - X_{II}) = 0.29496 .$$

This equation can be solved by using a computer, or by substituting gradually for X_{II} as follows:

for $X_{II} = 0.5$: $-\frac{\left[-11.0625 (1 - 0.5) + 7.92366\right]}{0.5} \ln 0.5 = 3.3165 ;$

for $X_{II} = 0.3$; $- \frac{\left[-11.0625\,(1-0.3) + 7.92366\right]}{0.3} \ln 0.7 = 0.2139$;

for $X_{II} = 0.305$: $- \frac{\left[-11.0625\,(1-0.305) + 7.92366\right]}{0.305} \ln 0.695 = 0.28061$;

for $X_{II} = 0.306$: $- \frac{\left[-11.0625\,(1-0.306) + 7.92366\right]}{0.306} \ln 0.694 = 0.29397$;

for $X_{II} = 0.3061$: $- \frac{\left[-11.0625\,(1-0.3061) + 7.92366\right]}{0.3061} \ln 0.6939 = 0.29534$;

and, after plotting the results of the last three equations versus the X values, as shown in Figure 3, it is seen that the calculated value of 0.29496 corresponds to $X_{II} = 0.30607$.

The changes in the composition of the liquid (N_{2x}), and the changes of the distribution coefficient (K_{calc}) as a function of the fraction solidified within range II are calculated according to Equations 2 and 3 as follows:

$$N_{2x} = {}^{1}\Big/\left[a + b\,X + c \ln (1 - X)\right] \tag{2}$$

$$K_{calc} = 1 + (A_2 - bX)\,N_{2x} \tag{3}$$

where:

$$a = \frac{1}{N_{22}} = \frac{1}{0.08} = 12.5,$$

$$b = \frac{A_2 - A_3}{X_{II}} = \frac{-11.0625 + 7.92366}{0.30607} = -10.255,$$

$$c = \frac{A_2(1-X_{II}) - A_3}{X_{II}} = \frac{-11.0625\,(1-0.30607) + 7.92366}{0.30607} = 0.807534.$$

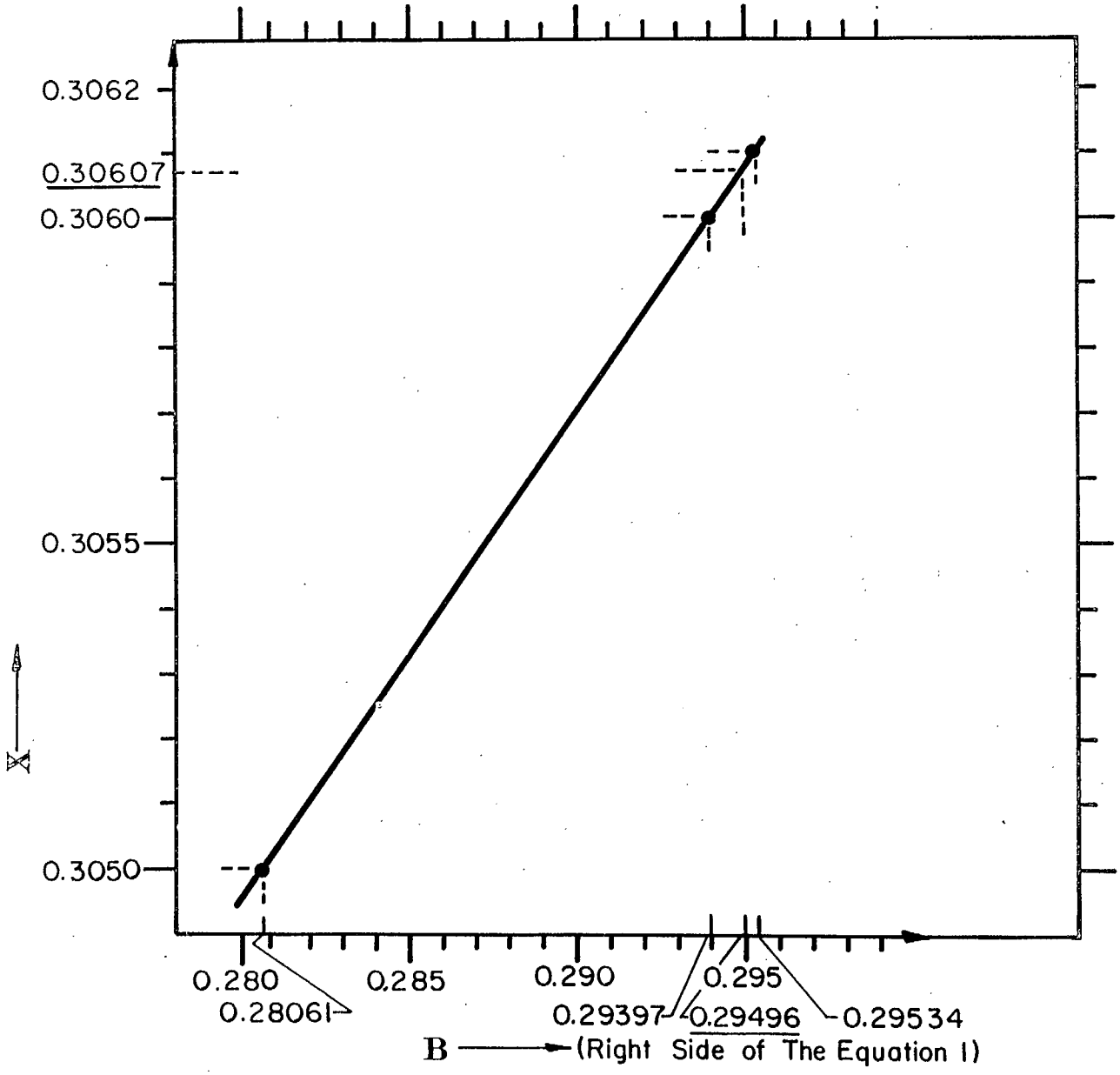

Figure 3. The graphical solution of the X values for Equation 1.

The changes of the N_{2x} and K_{calc} for calculated constants a, b, c, and for arbitrarily chosen X up to the final value 0.30607, are given in Table 3.

As the test for the accuracy of the calculation, the K_{calc} (0.126) should be in agreement with $K_{theor} = \frac{N_{(2)3}}{N_{23}} = \frac{0.0139}{0.1103} = 0.1260$.

The quantity of the liquid solidifying as the eutectic is calculated as an example for alloy 0.08 Mg_7Zn_3 from Equation 4 as follows:

$$X_E = (1 - X_{II})\ (1 - X_{III})\ \ldots\ (1 - X_{VIII})$$

$$= (1 - 0.30607)\ (1 - 0.37645)\ (1 - 0.54875)\ (1 - 0.37164)$$

$$(1 - 0.28026)\ (1 - 0.224255)\ (1 - 0.123854) = 0.060014 .$$

==

TABLE 3

The Changes in the Liquid Composition (N_{2x}) and the Distribution Coefficient (K_{calc}) Within Range II Calculated from Equations 2 and 3

$N_{2x} = 1/[a + bX + c\ln(1-X)]$					$K_{calc} = 1 + (A_2 - bX) N_{2x}$		
X	a	bx	cln(1-X)	N_{2x}	$A_2 - bX$	$(A_2-bX) N_{2x}$	K_{calc}
0.1	12.5	-1.02550	-0.085195	0.08700	-10.037	-0.88125	0.11875
0.2	12.5	-2.05100	-0.180161	0.09738	- 9.0115	-0.87754	0.12246
0.3	12.5	-2.56375	-0.232317	0.10305	- 8.4988	-0.87580	0.12420
0.30607	12.5	-3.13885	-0.295069	0.11030	- 7.9237	-0.87398	0.1260

APPENDIX B

Detailed Calculation of Solidification of Mg-6 wt% Zn Alloy

The calculation of the solidification process for the alloy containing 0.08 mole fraction of Mg_7Zn_3, within the limits of the change in the liquid composition from 0.08 up to eutectic (0.9366 mole fraction of Mg_7Zn_3), is as follows, according to Equations 6 to 11:

1) For X = 0,

$n_{2X} = N_{22} (1 - X) = 0.08 (1-0) = 0.08$

$n_{(2)x} = N_{22} - n_{2x} = 0.08 - 0.08 = 0$

$n_{1x} = 1 - X - n_{2x} = 1 - 0 - 0.08 = 0.92$

$n_{(1)x} = X - n_{(2)x} = 0.$

2) For range 1,

$X_1 = X_{II} = 0.30607$ (previous range II, Table 1)

$n_{2x} = N_{23} (1 - X_1) = 0.1103 \times 0.69393 = 0.07654$

$n_{(2)x} = N_{22} - n_{2x} = 0.08 - 0.07654 = 0.00346$

$n_{1x} = 1 - X_1 - n_{2x} = 1-0.30607 - 0.007654 = 0.686266$

$n_{(1)x} = X_1 - n_{(2)x} = 0.30607 - 0.00346 = 0.30262$

$\bar{N}_{(2)x} = \frac{n_{(2)x}}{X_1} = \frac{0.00346}{0.30607} = 0.01277.$

(Appendix B, concluded) -

3) For ranges 1 and 2,

$X_2 = 1 - (1-X_1)\ (1 - X_{III}) = 1 - (1-0.30607)$

$(1-0.37645) = 0.5673$

$n_{2x} = 0.1666\ (1-0.5673) = 0.072087$

$n_{(2)x} = 0.08 - 0.072087 = 0.007913$

$n_{1x} = 1 - 0.5673 - 0.072087 = 0.4248$

$n_{(1)x} = 0.5673 - 0.007913 = 0.55939$

$\bar{N}_{(2)x} = \frac{0.007913}{0.5673} = 0.01395$.

4) For ranges 1, 2 and 3,

$X_3 = 1 - (1-0.5673)\ (1-0.54875) = 0.804744$, etc., up to X_7.

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