MODELLING OF MICROSEGREGATION FOR TERNARY SYSTEMS : APPLICATION TO HOMOGENISATION OF AL-MG-SI ALLOYS

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ABSTRACT

Two numerical models have been developed for the quantitative prediction of microsegregation during solidification of ternary alloys. Both have been coupled with phase diagram calculations obtained with the ThermoCalc© software. The first model developed for globulitic grain structures considers the diffusion of the solute elements in one dimension (spherical geometry) with a front-tracking procedure for the solid-liquid interface. A two-dimensional microsegregation model based upon an explicit finite volume technique formulated for a regular hexagonal grid has also been developed for globular-dendritic grain structures. The discrete solid/liquid interface and the associated solute flux condition are accounted for by special "interfacial" volume elements. Such a model allows to calculate in two dimensions the segregation patterns and the precipitation of eutectic phases. It will be used to predict the homogenisation treatment of low-concentration alloys such as Al-1%Mg-1%Si.

1. INTRODUCTION

Segregation phenomena during solidification of metallic alloys are complex and have been the subject of many investigations (see Refs. in [1]). They can occur at two different scales. At the scale of the microstructure, the solidification path is associated with the partitioning and diffusion of the various solute elements in the liquid and solid phases (microsegregation). At the scale of the whole process, macrosegregation is mainly the result of transport of solute elements by convection.

The micro-macrosegregation phenomena are initiated in the mushy solid-liquid zone. Depending on the inoculation conditions, this region is made out of dendritic or globulitic grains [2]. In the first case, the grain size is fairly large with respect to the solute layer thickness and the internal structure of each grain is made out of well-developed dendrites. For globulitic grains, the solute layers that develop around the grains overlap at an early stage of growth and thus the spherical shape of the grain remains nearly stable. Intermediate globular-dendritic structures are frequently encountered in DC casting of aluminium alloys.

The modelling of micro-macrosegregation phenomena occurring in solidification processes requires to couple the macroscopic transport equations (mass, momentum, energy, solute) with a microscopic model describing the partitioning of the solute elements. In most cases, the complex three-dimensional shape of the solid-liquid interface is replaced by a one-dimensional (1D) microsegregation model. For dendritic structures, the size of this 1D volume element corresponds to the secondary dendrite arm spacing whereas for truly globulitic structures, it is simply given by the final grain radius. For intermediate globular-dendritic structures, however, this volume element is ill-defined and the precipitation of eutectic phases does not correlate to the dendrite arm spacing nor the grain size in any straight forward manner.

This contribution presents a 1D and a 2D model of microsegregation for multicomponent systems. The first model is aimed at being coupled with Finite Element (FE) macrosegregation calculations [1], whereas the second one addresses the questions of the repartition of eutectic phases in globular-dendritic aluminium alloys and of the subsequent homogenisation treatment.

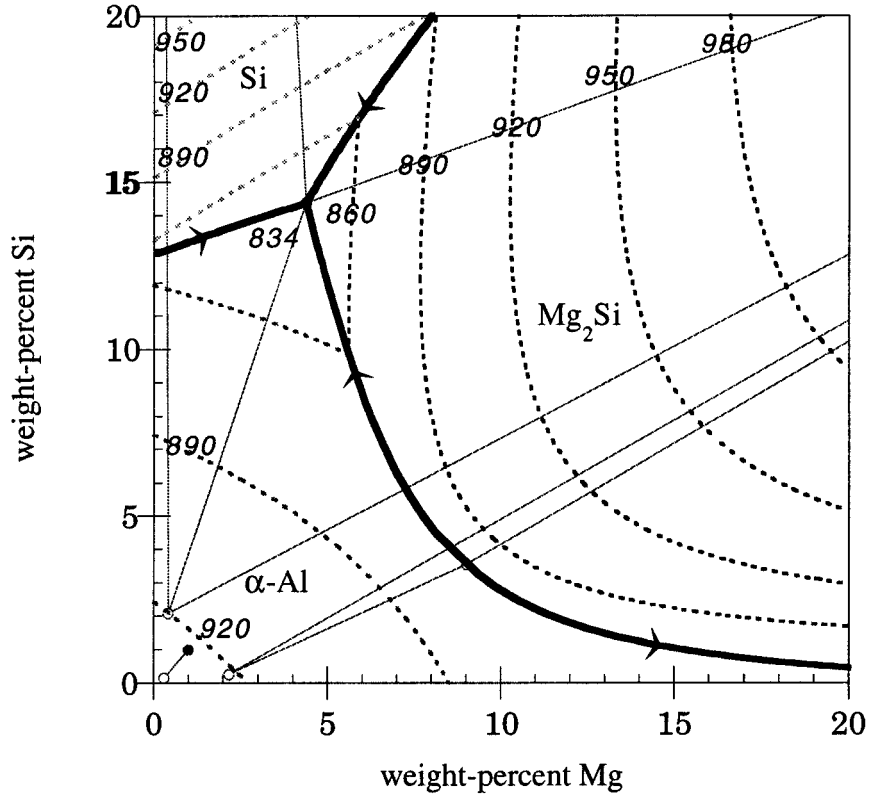

Fig. 1 Aluminium rich corner of the Al-Si-Mg phase diagram as calculated with ThermoCalc©.

2. PHASE DIAGRAM

The aluminium-rich corner of the ternary Al-Mg-Si phase diagram is shown in figure 1. It has been calculated with ThermoCalc© [3]. The dotted lines and the associated numbers correspond to a few liquidus isotherms for the α-Al, Mg_2Si

and Si regions. These three regions are separated by monovariant lines (thick lines) which meet at a ternary eutectic point (834 K). The small filled circle at the bottom left corner would correspond to the nominal concentration of an Al-1%Mg-1%Si alloy. A small segment links this concentration in the liquid phase with that of the solid that is in equilibrium (tie-line). Similarly, a concentration on a monovariant line for the liquid phase is in equilibrium with two concentrations in the corresponding solid phases (e.g., α-Al and Mg_2Si). This defines a triangle of tie-lines for these three phases in equilibrium. Please note that in this enlarged view of the phase diagram, two sides of such a triangle are nearly parallel. At the ternary eutectic point, six tie-lines link the concentrations in the four phases at equilibrium (only five tie-lines are visible in figure 1).

The information provided by such computed ternary phase diagrams must be linked with diffusion models in order to predict the solidification path of the alloy. In the case shown in figure 1, it is expected that the system will first solidify as a primary α-aluminium phase until the monovariant line $(l + \alpha) \leftrightarrow (l + \alpha + Mg_2Si)$ is reached. Because this line exhibits a saddle point, the end of solidification could be towards the "right" (continuous precipitation of Mg_2Si) or towards the "left", in which case some silicon might precipitate at the ternary eutectic point.

1D AND 2D MICROSEGREGATION MODELS

The solidification of the primary phase (e.g., aluminium in an Al-1%Mg-1%Si alloy) is emphasised in the present study. For a multicomponent alloy made out of N_c solute elements, the basic equations that have to be solved at the scale of the microstructure for the liquid and solid phases (ν = l, s) are as follows :

$$\text{div}\left(D_i^\nu \, \mathbf{grad}\, c_i^\nu\right) = \frac{\partial c_i^\nu}{\partial t} \qquad \text{for } i = 1, N_c \text{ and } \nu = l, s \qquad (1)$$

where c_i^ν is the volumetric concentration of solute element i in phase ν and D_i^ν is the corresponding diffusion coefficient . At the limit of the domain, the assumption of a closed system can be made if macrosegregation is neglected. A flux of solute can also be introduced for an open system [1]. At the moving solid-liquid interface, a solute flux balance has to be satisfied :

$$\left[D_i^l \frac{\partial c_i^l}{\partial n} - D_i^s \frac{\partial c_i^s}{\partial n}\right]^* = - v_n^{s/l} \left[c_i^l - c_i^s\right]^* \qquad \text{for } i = 1, N_c \qquad (2)$$

where the index "*" denotes entities taken at the s/l interface, **n** is the normal to the interface pointing in the liquid and $v_n^{s/l}$ is the normal velocity of the interface. In other words, the sum of the solute fluxes at the interface must be equal to the jump of concentrations multiplied by the normal velocity. The concentrations at the s/l interface are linked to the phase diagram via the relationships :

$$c_i^{s\,*} = c_i^s\left(c_1^{l\,*}, c_2^{l\,*}, \ldots, c_{N_c}^{l\,*}\right) \qquad \text{for } i = 1, N_c \qquad (3)$$

$$T = T_L\left(c_1^{l\,*}, c_2^{l\,*}, \ldots, c_{N_c}^{l\,*}\right) \tag{4}$$

The first N_c equations correspond to the tie-lines of the s/l interface, whereas eq. (4) is the equation of the liquidus surface. Providing the temperature is uniform at the scale of the microstructure and known as a function of time, eqs. (1-4) form a set of equations which allows to calculate the solidification path of the primary phase up to a eutectic reaction.

1D model

Since the details of the 1D model can be found in [1], they will not be repeated here. It will just be reminded that the solidification of the primary α-phase is calculated until a monovariant line is reached. The moving α/l interface is tracked using a Landau transformation of coordinates. The diffusion equations in the solid and liquid phases are solved in the transformed (fixed) mesh with the help of a fully-implicit finite difference (FD) scheme. Once a monovariant line is reached, the remaining liquid is transformed into solid phases according to a Scheil-Gulliver approximation [4]. This law gives the amount of the various phases (Mg_2Si, Si) which precipitate during the binary/ternary eutectic reactions.

2D model

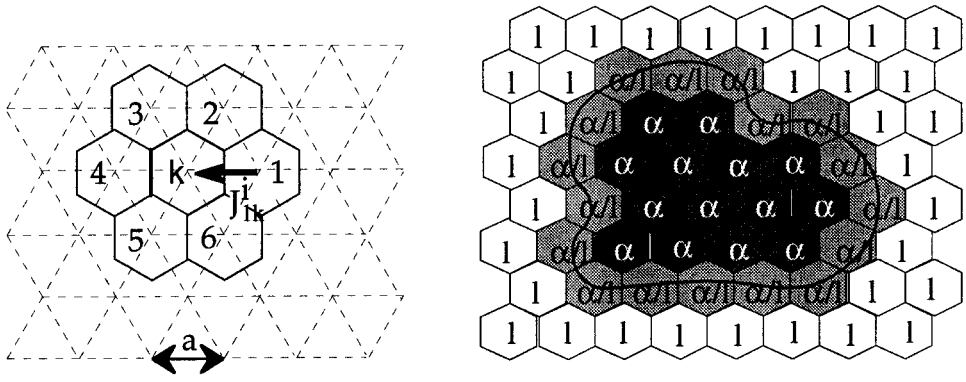

Fig. 2 *Enmeshment used in the 2D microsegregation model.*

The 2D model is derived from that developed by Jacot and Rappaz for the solid-state transformation in steels during heating [5]. A 2D domain representative of the microstructure is subdivided into a regular arrangement of hexagonal cells (see figure 2). The cells can be liquid (l), solid (α) or "mushy" (α/l). For each of them, the volume fraction of liquid, f_l, and the concentrations c_i^l are defined as independent variables. An explicit Finite Volume scheme is used to solve eq. (1) and to deduce the variations of concentrations in each cell. For the mushy cells, these variations correspond to an average over the liquid and solid phases. Since the phase diagram conditions (eqs. (3) and (4)) must be respected, the variations of the average concentrations in these cells are converted into a variation of the fraction of liquid using a local lever-rule approximation. Once a mushy cell is fully solid, it becomes an α-cell and new interfacial α/l cells are created around it (i.e., liquid

neighbour cells become interfacial ones). It was shown that such a procedure diffuses the interface over one mesh element but satisfies the solute flux balance (eq. (2)) and the equilibrium conditions (eqs. (3) and (4)). Further details of this algorithm can be found in Ref. 5.

RESULTS

Figure 3 shows the Si concentration profiles at various times of solidification for an Al-1%Mg-1%Si alloy. Spherical coordinates (i.e., fully globulitic growth) were used in these computations and the final grain radius, r_o, was assumed to be 50 μm. The other parameters used in this computation are listed in the figure caption. As can be seen, the solute is almost totally mixed with the liquid phase, owing to rapid diffusion in this phase. The last profile shown corresponds to the time at which the monovariant line ($l + \alpha + Mg_2Si$) is reached. Some back-diffusion can be noticed in the primary phase (i.e., the concentration profile in the solid is not totally "frozen" but evolves with time).

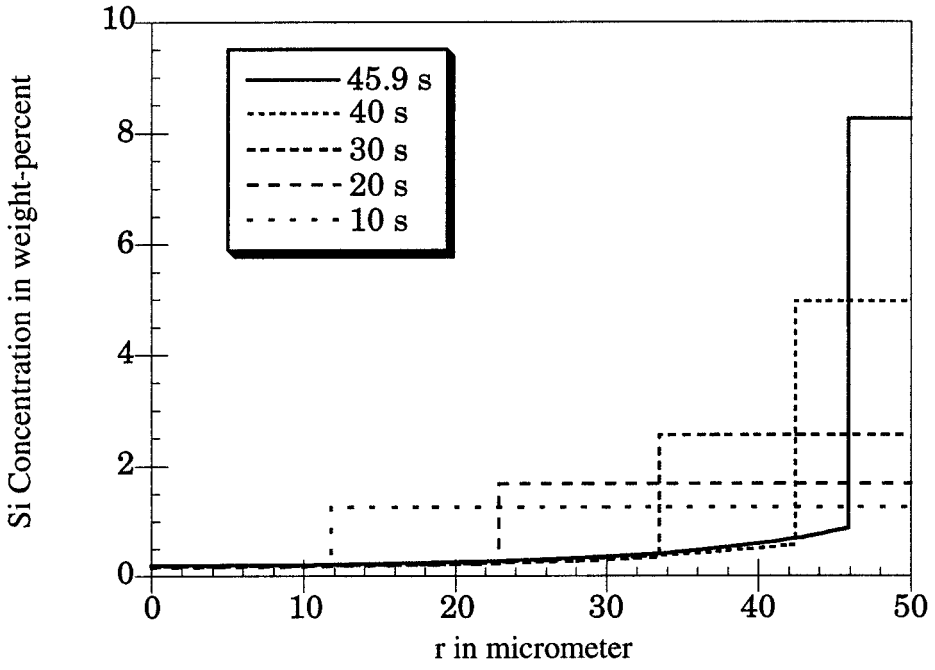

Fig. 3 Evolution of the silicon solute profile in a Al-1%Mg-1%Si alloy solidified at a heat extraction rate of -9.10^3 W/kg, L = 388 kJ/kg, c_p = 1080 J/kgK, D_l (Si) = $2.45 \cdot 10^{-8}$ m^2/s, D_s (Si) = $4.91 \cdot 10^{-12}$ m^2/s, D_l (Mg) = $9.72 \cdot 10^{-9}$ m^2/s, D_s (Mg) = $4.5 \cdot 10^{-12}$ m^2/s.

The associated cooling curve, T(t), and evolution of the volume fraction of solid, $f_s(t)$, are shown in figure 4. In this geometry, $f_s(t)$ is simply given by $(r(t)/r_o)^3$, where r(t) is the position of the solid-liquid interface shown in figure 3. Since the heat extraction rate, $\dot{h}$ was given instead of the temperature, a heat balance was made at the scale of the grain to close the problem :

$$\dot{h} = c_p \dot{T} - L \dot{f}_s \qquad (6)$$

where c_p and L are the volumetric specific heat and latent heat, respectively. (The dot over the variables indicate time derivatives). The small squares in figure 4 correspond to the primary phase solidification, whereas small circles and diamonds are used for the binary and ternary eutectics, respectively.

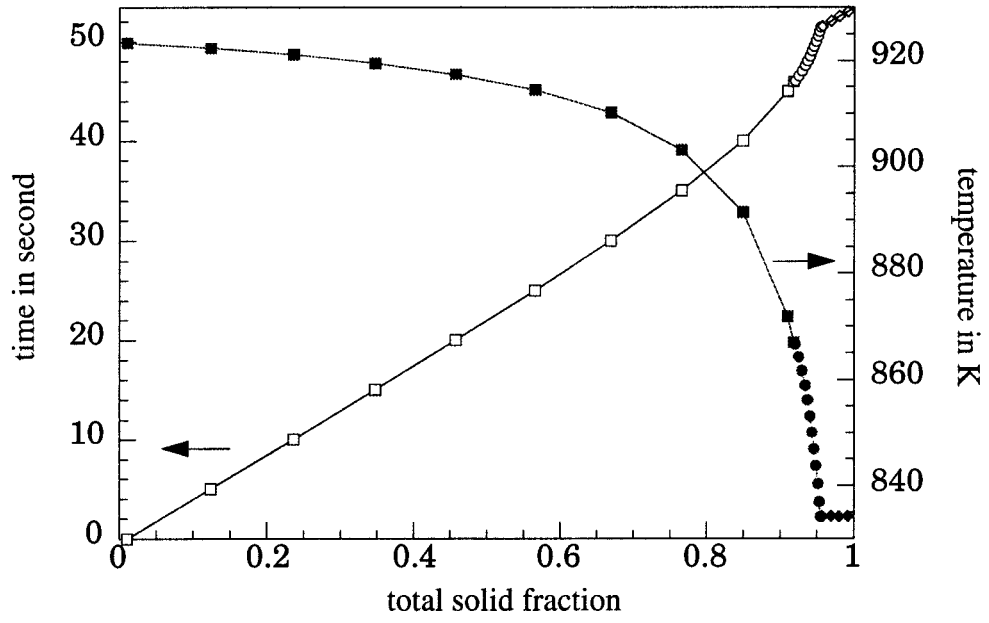

Fig. 4 *Evolutions of the fraction of solid and temperature as a function of time for the alloy and the conditions of figure 3.*

Figure 5 shows a result of the 2D microsegregation model for the same alloy. This map corresponds to the volume fraction of solid, at a time just before the monovariant line is reached (see figure 1). White is fully solid and black is liquid. The size of the domain is 500 μm x 500 μm and 16 grains were randomly nucleated within the domain at the liquidus temperature. Thus, the average grain size in figure 5 is of the order of that used in figure 3. 10'000 cells were used in this computation. It is interesting to note the inhomogeneous repartition of the last liquid in figure 5 (dark zones) which ultimately will become the intergranular eutectics. Such a map can be qualitatively compared with the experimental micrograph shown in figure 6.

In the figure 7, the average concentration within each cell was used. The concentration profiles show that there are some small variations of concentration in the solid, as already noticed in figure 3. The sharp peaks seen in this figure correspond to the last remaining liquid (or mushy) pockets in the microstructure.

CONCLUSION

A 1D microsegregation model coupled with phase diagram computations has been developed. The fast front-tracking algorithm can give accurate results with only a few nodal points in each phase [1]. It could therefore be used easily as a subroutine

coupled with the FE solution of the macroscopic continuity equations in order to predict micro-macrosegregation in real alloys.

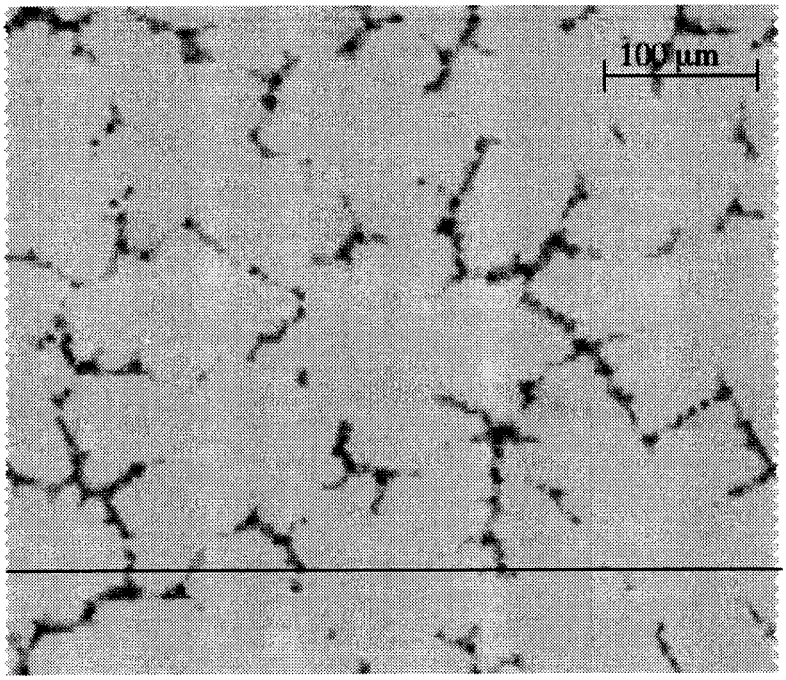

Fig. 5 *Volume fraction map as calculated with the 2D microsegregation model, just before the monovariant line is reached. The alloy and parameters used in the calculation are the same as those of figure 3 except for a cooling rate of -1 K/s .*

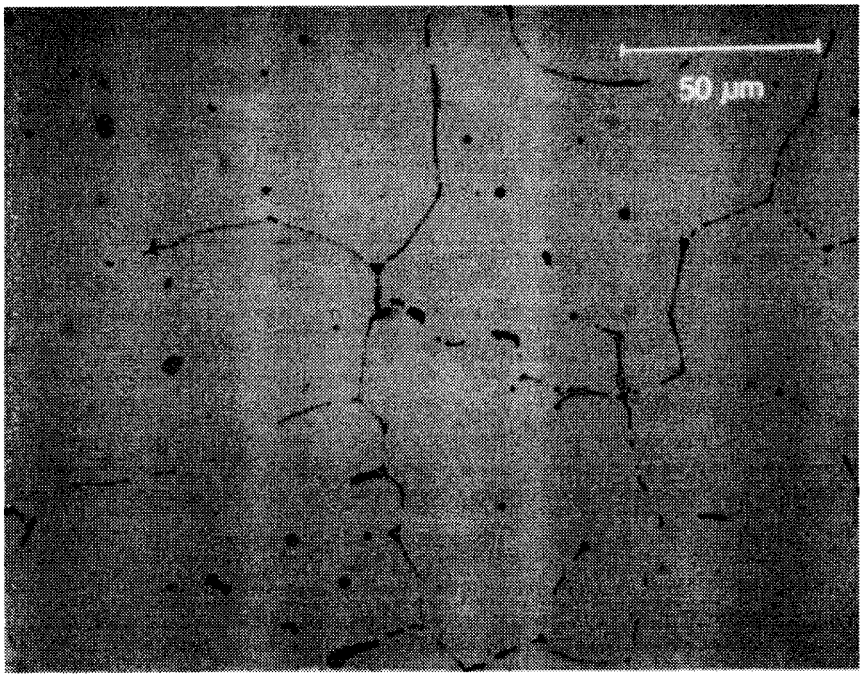

Fig. 6 *Repartition of eutectic in a micrograph of a Al-1%Mg-1%Si alloy.*

On the other hand, a 2D model which spreads the solid-liquid interface over one mesh has been adapted to the case of multicomponent alloys. It can predict the repartition of intergranular eutectic and will be used in further works to study the

homogenisation process. Prior to that, the curvature undercooling of the interface will be introduced in the calculation in order to include dendrite arm coarsening and to obtain more realistic microstructures.

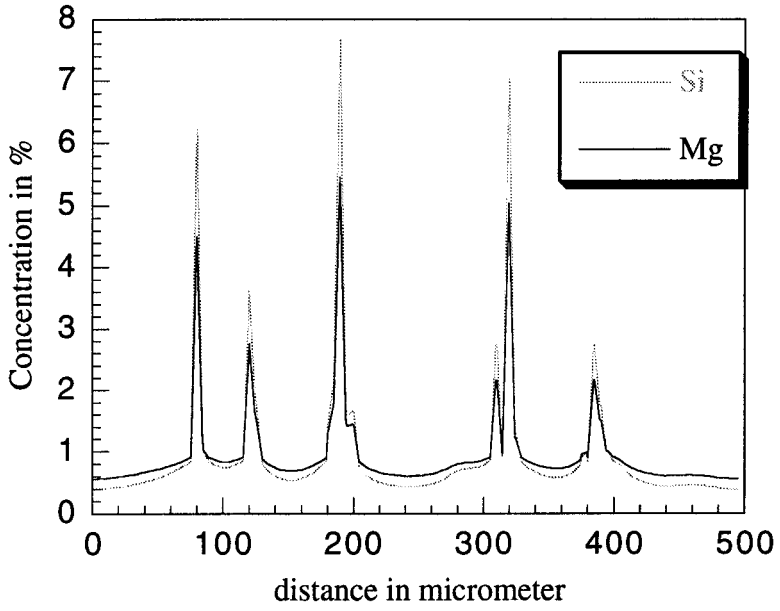

Fig. 7 *The solute profiles along the horizontal line drawn in figure 5.*

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