Determination of the cast structure parameter on the basis of micro-segregation analysis

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ABSTRACT. *The paper deals with the new method for estimation of the average dendrite arm spacing on the basis of parameters of solidification of the relevant metallic alloy and of the theory of physical similarity. The following parameters determine dendritic structure of cast metallic alloys: diffusion coefficient of elements in solid D_S, partition coefficient of elements k, local solidification time Θ, rate of crystallization w and chemical heterogeneity of the given alloy, quantified usually with maximum c_{max}, and initial c_0 concentration of elements in the given area of alloy. Verification of the method was carried out both theoretically and experimentally. An average secondary dendrite arms spacing has been calculated as a function of the cooling rate according to relations given in literature. The dendrite arm spacing was also estimated metallographically for nine samples taken from cross section of a low-carbon continuously cast steel. It was found out that for short local solidification times the average dendrite arm spacing determined with use of the new method was lower than values calculated according to literature or determined metallographically. However, calculation for long local solidification times with use of the new method corresponded very well with both calculated and measured values.*

INTRODUCTION

Character and quality of cast micro-structure influence tendency of metallic materials to cracks and fractures. The large dendrite arm spacing may increase the interdendritic areas and concentrations of solute elements in the interdendritic liquid. A coarse structure also tends to increase the micro-segregation of solute and impurity elements and, consequently, the related cracking susceptibility of the steel.

To estimate the cracking condition in continuously cast steels, a new model for critical fracture stress given from the measured critical strain has been proposed in [1], which can take into account the brittle temperature range and strain rate. The brittle temperature range needs to be computed from the steel composition, cooling rate, and dendrite arm spacing; the brittle temperature range is calculated with micro-segregation

analysis. The effect of dendrite arm spacing on hot tearing susceptibility during directional solidification of a directionally solidified nickel alloy was explored in [2]. It was found that smaller dendrite arm spacing does reduce the hot tearing tendency. The study [3] reveals the micro-mechanisms of fatigue crack nucleation and growth in a commercial high-pressure die cast automotive AZ91E-T4 Mg component. It was found out that secondary dendrite arm spacing and grain size are micro-structural features that have been shown to influence the overall fatigue life of cast materials.

It follows from the above mentioned that the structural parameter characterizing distances between dendrites (dendrite arm spacing) is one of the most important for description and prediction of crack mechanism in metallic materials.

The following quantities have particularly important influence on dendritic structure of cast materials: - rate of crystallisation w (ms^{-1}); - local time of solidification Θ (s); i.e. time, during which the considered area of solidifying body has a temperature between that of solidus and liquidus; - effective partition coefficient k_{ef} (-) of elements, - diffusion coefficient D_S (m^2s^{-1}) of segregated element in solid phase; - chemical heterogeneity of element, usually assessed by means of mean concentration of element in the alloy C_m (wt.%), its standard deviation σ_C (wt%), or possibly its maximum concentration of the element in the given area (C_{max}). Dendritic structure is the most frequently characterised quantitatively by the average dendrite arms spacing L (m).

Aim of this paper is to indicate that it is possible to (approximately) estimate the average dendrite arms spacing, i.e. average distance between dendritic axes, with use of parameters of solidification of the relevant metallic alloy, diffusivity, effective partition coefficient and chemical heterogeneity of the given alloy.

THEORETICAL BACKGROUND - RELATIONS BETWEEN PARAMETERS INFLUENCING DENDRITIC STRUCTURE

Relation between diffusion coefficient D_S of the element in a solid phase, local solidification time Θ, dendrite arms spacing L, local rate of crystallisation w, mean concentration of constitutive element C_m, including standard deviation of this concentration σ_C and effective partition coefficient of the element between solid phase and melt k_{ef} can be, on the basis of existing level of knowledge and findings, considered to be dominant with respect to forming of dendritic structure of metallic alloy. Out of the seven quantities specified above the mean concentration of element measured in the given area, and its standard deviation, define the index of chemical heterogeneity I_H:

$$I_H = \frac{\sigma_C}{C_m} \tag{1}$$

In this way the number of quantities is reduced by one quantity and it is possible to describe formally relation between them by the following equation:

$$f(D,\Theta,L,w,I_H,k_{ef}) = 0 \tag{2}$$

On the basis of theory of physical similarity it is possible to substitute the Eq. (2) with use of dimensional analysis of functions between dimensionless criteria of similarity. It is possible to find the necessary criteria of physical similarity and their number with use of the π - theorem. This is possible with use of matrix of dimensions of the considered six quantities and by its subsequent transformation to the matrix of criteria, in which the considered quantities appear after this transformation. Matrix of dimensions-A comprises in the given case 6 quantities, general dimensions of which can be expressed by 3 basic dimensions (Table 1).

Table 1. Matrix of dimensions (A) and matrix of dimensionless groups (B)

Matrix A							Matrix B						
Dimension	***D***	***Θ***	***L***	***w***	I_H	k_{ef}	**Criterion**	***D***	***Θ***	***L***	***w***	I_H	k_{ef}
m	2	0	1	1	0	0	$\pi_1 = \alpha$	1	1	-2	0	0	0
s	-1	1	0	-1	0	0	$\pi_2 = Th$	0	1	-1	1	0	0
-	0	0	0	0	1	1	π_3	0	0	0	0	1	-1

In accordance with the π - theorem mentioned above it is possible to substitute the relation between the mentioned six variable quantities by three dimensionless criteria of similarity (number of dimensionless criteria equals number of variable minus number of basic dimensions, i.e. 6 – 3 = 3). In this way we obtain the matrix of criteria B in the form given in Table 1.

We can see that it is possible to substitute the original function expressed by Eq. (2) by the function between three criteria of similarity

$$F(\pi_1,\pi_2,\pi_3) = F(D_S\Theta/L^2, w\Theta/L,\ I_H/k_{ef}) = 0 \tag{3}$$

The first criterion $\pi_1 = D_S\Theta/L^2$, which expresses Fourier's number (criterion) for mass transfer, is frequently used in the models of solidification (micro-segregation) and has the sign α. This dimensionless criterion is used in models describing segregation of elements at crystallisation of metallic alloys. This criterion expresses in general a relation between the rate of mass transfer in a solid body (or area), with use of physical properties and dimensions of the considered body (area). In the given case, i.e. in dendritic structure this criterion gives information about intensity of diffusion processes in solid phase at crystallisation between liquidus and solidus, and in great extent also information about intensity of these processes during cooling down below the solidus temperature.

The second dimensionless criterion π_2 in the Eq. (3) has in its numerator a product of local rate of solidification and local solidification time between liquidus and solidus temperature, when solid and liquid phases coexist in the mixture (mushy zone). The product in the numerator $w\Theta$ is related to the average dendrite arms spacing L. This

criterion is called a Thomson's criterion Th, Th = wΘ/L. It represents a universal criterion of kinetic similarity of phenomena. This criterion can be used for description of unsteady flow of melt during solidification between the liquidus and solidus temperature within the frame of dendrites with characteristic dendrite arms spacing L.

The third criterion $\pi_3 = I_H/k_{ef}$ in the Eq. (3) represents a simplex, expressed by relation of the index of chemical heterogeneity of segregated element within the frame of dendrite, to the effective partition coefficient of the element between solid and liquid phase during crystallization.

DENDRITE ARMS SPACING ESTIMATION AND ITS VERIFICATION

On the basis of evaluation of the set of measurements of chemical heterogeneity of alloying and impurity elements in selected areas of specimens taken from continuously cast steel slab it was shown, that it was possible to determine from the measured concentrations of elements the following quantities [4]:

a) index of chemical heterogeneity I_H of each of the measured element (at the same time it is possible to measure in each point of the area a concentration of 7 to 10 elements),

b) it is possible to determine for each measured element also effective partition coefficient k_{ef}.

In this way it is possible to determine relation between the index of heterogeneity of elements I_H and their effective partition coefficients k_{ef}, i.e. values of criterion π_3 (Eq. (3)).

It has also been verified that it was possible to determine by statistical processing of concentrations of elements measured in the given area of structure the most probable shape of distribution curve of dendritic segregation of the relevant element [5]. The distribution curve expresses a probable distribution of concentration of this element in the dendritic structure. If we know the shape of dendritic curve of the relevant element, it is then possible to determine for model of micro-segregation of the element during solidification a value of the first dimensionless criterion in the matrix B (Table 1), i.e. $\pi_1 = \alpha = D_S\Theta/L^2$. In the works [4, 5] the Brody-Flemings model was chosen as suitable model.

However, by modification of this criterion we obtain the relation

$$L^2 / \Theta = D_S / \alpha = A\,. \tag{4}$$

Constant value of the parameter A (m^2s^{-1}) enables estimation of average dendrite arms spacing in accordance with the simple relation

$$L = \sqrt{A\Theta} \tag{5}$$

as a quantity, which is directly proportional to the square root of the local solidification time. However, local solidification time Θ represents a parameter, the value of which is determined only with great difficulty. One of the ways for its determination is differential thermal analysis (DTA), which can be used only for laboratory measurements. Another way, which can be used, namely for large bodies, is numerical modelling of temperature field of the given body (see e.g. the works [6] and [7]).

For verification of the described approach to (approximate) estimation of dendritic structure of continuously cast steel slab we have used already previously published measurements of chemical micro-heterogeneity of elements, their partition coefficients and also local solidification times, calculated with use of 3-D numerical model of non-stationary temperature field of solidified slab [9,10].

Chemical composition of steel (in wt.%): 0.14C; 0.75Mn; 0.23Si; 0.016P; 0.010S; 0.10Cr; 0.050Cu; 0.033Al_{total}. A transverse strip was cut from the cast slab after solidification and cooling, which was axially divided to halves. Altogether 9 samples were taken from one half, see Fig. 1. Concentration distribution of eight elements (Al, Si, P, S, Ti, Cr, Mn and Fe) was measured on each sample along the abscissa with length of 1000 μm. Spacing between the measured points was 10 μm. Analytical complex JEOL JXA 8600/KEVEX Delta V Sesame and method of energy dispersive analysis were used for determination of concentration distribution of elements. The work [8] presents results of measurements, including determination of values of effective partition coefficient, indices of chemical heterogeneity and determined values of dimensionless criteria α for all analysed elements on individual samples. The data are completed by values of diffusion coefficients in solid phase taken from literature and also by values of local solidification times determined on the basis of calculation from numerical model of non-stacionary temperature field of the considered steel slab.

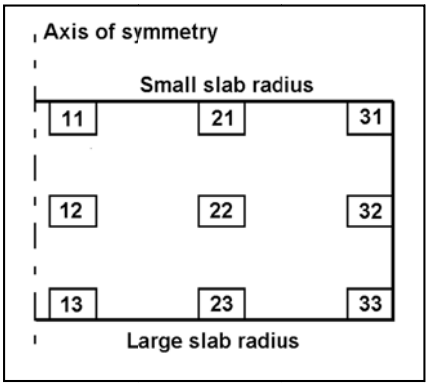

Figure 1. The scheme of sampling. Small and large slab radius corresponds to the small and large radius of a radial continuous-casting machine.

Data from the work [8], which were used for calculation of spacing between dendrite arms, are for clearness given in Table 2. These are values of dimensionless criterion α calculated from experimentally measured concentration data, including standard

deviation σ_α. It contains moreover values of diffusion coefficients in solid phase D_S and calculated constants A according to Eq. (4). Mean value of the constant A is $A = 632.3 \pm 372.4$ $\mu m^{-2}s^{-1}$. Values of spacing between dendrite arms L^{calc} were then calculated for individual samples according to Eq. (5). Local cooling rate v (Ks^{-1}) has been determined with use of numerical model of a 3D non-stationary temperature field. Values L^{calc} (as well as values $L^{calc}+\sigma_L$ and $L^{calc}+\sigma_L$) and values of cooling rate v for individual samples are given in Table 3.

The following two methods have been used for a basic verification of estimated values of dendrite arms spacing:

a) An average secondary dendrite arms spacing L^{lit} has been calculated with use of relation published in [9] as a function of the cooling rate v. Values L^{lit} for individual samples are given in Table 3.

b) A dendrite arms spacing has been determined on samples taken from a continuously cast slab, metallographically on planar polished sections. Dendritic structure of the samples was developed by chemical etching. Its pictures were then taken at magnification 20x and spacing between dendritic axes was measured on micro-graphs by linear method. Values of distances between axes in individual samples were determined as a mean from nine measurements. The values measured in this manner are in Table 3 marked as L^{mea}.

Table 2 Data for calculation of an average dendrite arms spacing

Parameter	Al	Si	P	S	Ti	Cr	Mn	Fe
$\alpha.10^2$	2.404	2.171	2.005	2.901	2.214	2.672	1.334	1.067
$s_\alpha \cdot 10^2$	0.607	0.642	0.498	0.506	0.224	0.964	0.599	0.512
$D_S \cdot 10^8$ (cm^2/s)	61.942	0.624	5.340	59.800	0.585	1.649	0.237	0.204
$A \cdot 10^7$ (cm^2/s)	257.66	2.874	26.633	206.14	2.642	6.171	1.776	1.912

Table 3 Dendrite arms spacing determined by two methods and by Eq. (5)

Sample	v (K/s)	Θ (s)	L^{lit}	$L^{mea} \pm \sigma_L$	L^{calc}	$L^{calc}+\sigma_L$	$L^{calc}-\sigma_L$
11	2.017	8.54	107.3	172 ± 12	73.5	92.6	47.1
12	0.035	357.34	565.3	407 ± 57	475.3	599.2	304.8
13	2.017	8.54	107.3	–*)	73.5	92.6	47.1
21	1.712	10.01	114.7	–*)	79.6	100.3	51.0
22	0.032	384.67	586.4	–*)	493.2	621.7	316.2
23	1.712	10.01	114.7	212 ± 11	79.6	100.3	51.0
31	2.008	8.18	107.5	183 ± 18	71.9	90.7	46.1
32	0.674	25.18	168.1	180 ± 16	126.2	159.1	80.9
33	2.008	8.18	107.4	172 ± 9	71.9	90.7	46.1

*Note: *) it was impossible to etch the dendritic structure, only the micro-structure of steel itself was etched*

DISCUSSION

It can be seen from comparison of dendrite arms spacing (Table 3), the values of which were determined in three different ways, that the values determined by calculation according to the relation given in [9] and determined by metallographic measurements (i.e. L^{lit} and $L^{mea} \pm \sigma_L$) are in relatively good correspondence. It must be stressed that the relation in [9] is determined for calculation of secondary dendrite arms spacing. Values of spacing between dendrite arms determined metallographically by linear method comprise logically dendrite arms of various orders. The mean value of dendrite arms spacing determined with use of the Eq. (5) deviates for short local solidification times towards lower values of L. However, calculation for long local solidification times with use of the Eq. (5) corresponds very well with both previous measurements (i.e. with the values of L^{lit} and $L^{mea} \pm \sigma_L$).

The mean error of the arithmetic mean of the constant A is rather big, relative error is 58.9 %, and as it follows from Table 2 this error is caused by big differences of the constant A between the two groups of elements. On one side there are the elements – Si, Ti, Cr, Mn and Fe, for which the value of the constant A is in units cm^2/s, and on the other side there are three elements – Al, P and S, for which the constant A has a value higher by one or even two orders, as compared with the previous group of five elements. At the same time it is remarkable that elements, such as Al, S and P have under otherwise identical conditions higher diffusivity (D_S) than the remaining group of elements (i.e. Si, Ti, Cr, Mn and Fe).

It is, however, possible to determine the values of the constant A retrospectively from the values measured metallographically with use of Eq. (5). We will get from the six measured values L^{mea} given in Table 3 with use of the Eq. (5) the mean value $A = (290.3 \pm 163.3) \cdot 10^{-7}$ $cm^{-2}s^{-1}$, which is very close to the value of the constant A for aluminium (see Table 2).

It follows from the above that the proposed method of (approximate) evaluation of spacing between dendrite arms is suitable for application for slow solidification of large metallic bodies.

CONCLUSIONS

The paper describes approach to a qualified estimation of the average dendrite arms spacing, which enables a comparatively rapid obtaining of basic data about dendritic structure of steel and other dendritically solidifying metallic alloys, under the following presumptions:

a) quantitative data about chemical micro-heterogeneity of elements (the concentration profiles) in the given area of the body are available, and

b) the local solidification time in the same area of the body is known.

This proposed method was used in the paper for calculation of spacing between dendrite arms for the samples taken from cross-section of continuously cast slab. On the basis of concentration profiles of eight elements and values of local solidification times

calculated with use of numerical model of non-stacionary temperature field the values of dendrite arms spacing were determined for individual samples. The Eq. (5) given in this paper has been found suitable for this calculation.

The calculated values were compared with metallographically measured values of dendrite arms spacing and with values calculated according to the relation in [9]. It was established that the proposed method of (approximate) estimation of dendrite arms spacing is suitable for application for slow solidification of large metallic bodies. It gives lower values of spacing between dendrite arms for short solidification times.

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