

Comment on Stefanescu, D.M.; Alonso, G.; Suarez, R. Recent Developments in Understanding Nucleation and Crystallization of Spheroidal Graphite in Iron-Carbon-Silicon Alloys. Metals 2020, 10, 221

Jacques Lacaze, Manuel J Castro-Roman

▶ To cite this version:

Jacques Lacaze, Manuel J Castro-Roman. Comment on Stefanescu, D.M.; Alonso, G.; Suarez, R. Recent Developments in Understanding Nucleation and Crystallization of Spheroidal Graphite in Iron-Carbon-Silicon Alloys. Metals 2020, 10, 221. Metals, 2020, 10 (4), pp.471. 10.3390/met10040471. hal-02539839

HAL Id: hal-02539839 https://hal.science/hal-02539839

Submitted on 10 Apr 2020 $\,$

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Comment



Comment on Stefanescu, D.M.; Alonso, G.; Suarez, R. Recent Developments in Understanding Nucleation and Crystallization of Spheroidal Graphite in Iron-Carbon-Silicon Alloys. *Metals* 2020, *10*, 221.

Jacques Lacaze ^{1,*} and Manuel J. Castro-Roman ²

- ¹ Centre Inter-universitaire de Recherche et d'Ingénierie des Matériaux (CIRIMAT), Université de Toulouse, BP 44362, 31030 Toulouse, France
- ² Center for Research and Advanced Studies of the National Polytechnic Institute, Saltillo Campus, Av. Industria Metalúrgica, 1062, Parque Industrial Saltillo-Ramos Arizpe, Ramos Arizpe 25900, Coahuila, México (CINVESTAV Saltillo); manuel.castro@cinvestav.edu.mx
- * Correspondence: Jacques.lacaze@ensiacet.fr; Tel.: +33-534323415

Received: 11 March 2020; Accepted: 15 March 2020; Published: 3 April 2020

In an article published in this journal in February 2020, Stefanescu et al. reviewed recent developments in understanding nucleation and growth of spheroidal graphite in cast iron [1]. The intention of the present comment is not to discuss the merits and limits of this highly documented paper in general, but to correct erroneous statements made by the authors in their Section 5.1. One of the approaches reviewed in this section is the work by Theuwissen et al. [2], who suggested that the 2-D poly-nucleation-growth (PNG) mechanism applied by Amini and Abbaschian [3] to describe the formation of polyhedral graphite plates in Ni–C alloys might also be operational for flake and spheroidal graphite in cast iron melts. This hypothesis has been supported by the analytical calculations of Lacaze et al. [4], and it is this latter work which is severely criticized by Stefanescu et al. in section 5.1 of their paper [1]. In the following, we detail that the supposed numerical incongruities and seemingly shortcomings of the work by Lacaze et al. [4] are not real, but are based on a severe misinterpretation of the analytical model parameters and results.

The 2-D nucleation-growth model for spheroidal graphite by Lacaze et al. [4] expresses the overall growth rate of a nodule from the liquid, GPNG, as (Equation (14) in [4]):

$$G_{PNG} = \mathbf{a} \cdot \left(\frac{\pi}{3} \cdot \mathbf{J}_{a} \cdot (\mathbf{V}_{1})^{2}\right)^{1/3}$$
(1)

where:

- a is the height of the nuclei, which is set equal to the distance between basal planes of graphite.
- J_a is the nucleation rate of new growth units at the outer surface of the nodule and is described by a classic approach for 2D nucleation [5].
- V₁ is the lateral spreading rate of the growth units.

A first approach to express V₁ was to consider a general kinetic law derived by Cahn et al. [6], which leads to the following equation (Equation (17) in [4]):

$$G_{PNG} \approx 0.05 \cdot \left(\Delta T\right)^{5/6} \cdot \beta \cdot \left(2 + \xi^{-1/2}\right)^{2/3} \cdot \exp\left(-\frac{7200 \cdot \xi}{\Delta T}\right) \ m.s^{-1}$$
(2)

where ΔT is the undercooling with respect to the graphite liquidus, β denotes a structural parameter and ξ is the diffuseness of the interface as defined by Cahn et al. [6].

The structural factor was set to 1, as it should be for single atoms (as opposed to molecules) [5], and ξ was varied between 0.1 and 1 which is its maximum value (sharp interface). This gave the curves plotted with dotted lines in Figure 1a.

A second way to express V₁ was based on carbon diffusion from the liquid to the growing units. The solution proposed was according to Bosze and Trivedi [7], which is more precise than the approach used by Amini and Abbaschian [3]. This led to the following equation (numbered 29 in [4]):

$$G_{PNG} = 2.6 \cdot 10^{-11} \cdot (\Delta T)^{17/6} \cdot (\beta)^{1/3} \cdot \exp\left(-\frac{7200 \cdot \xi}{\Delta T}\right) \text{ m.s}^{-1}$$
(3)

The structural factor was set to 1 as before, and the same values were used for the diffuseness, giving the solid curves in Figure 1a.

Lacaze et al. calculated growth rates in the range of undercooling of 0–250 °C in agreement with the fact that experiments on primary graphite growth from the liquid are carried out on melts having been carbon saturated at high temperature—typically 1350–1550 °C—to get a large enough temperature interval before bulk eutectic solidification occurs, following in this many previous authors [3,8,9].

The first criticism of Stefanescu et al. is the choice of the range of the undercooling ΔT . The authors claim that an "undercooling higher than about 50 K will be conducive to metastable structure in Fe-C-Si alloys". Stefanescu et al. [1] obviously misunderstood the undercooling as a technical undercooling evaluated with respect to the eutectic instead of with respect to the graphite liquidus.

Then, referring to the figure reproduced here in Figure 1a, Stefanescu et al. "also notice some numerical incongruities: At a reasonable undercooling of 25 K (thin vertical line on the figure) it will take 3 hrs". These authors thus also mixed up the plots, as our final discussion was dealing with results calculated with Equation (3) which appear as solid lines in Figure 1a. To avoid any further misunderstanding, the curve calculated with Equation (3) and the selected value of 0.1 for diffuseness is reproduced in Figure 1b.

Finally, Stefanescu et al. claimed "Such unrealistic calculation results are not surprising given the number of parameters that have to be estimated (e.g., the height of the nucleating disk, diffuseness, the correction for the structural factors)". Thus, these authors wrongly accused us of using a high number of fitting parameters, while there was only a single one, the interface diffuseness, and the impact of this one has been discussed.

The appropriateness of our approach was checked against a few available experimental results [10] in the last section of our paper [4]. To sum up, we certainly think that analytical approaches could still be of some help for understanding metallurgical processes and in particular spheroidal graphite growth.



Figure 1. Overall growth rate of a graphite spheroid according to the PNG model as a function of the undercooling with respect to the graphite liquidus. The horizontal line represents the experimental value estimated by Amini and Abbaschian for the thickening rate of lamellar graphite (growth along the basal-c direction). ξ is the diffuseness of the interface. In (**a**) is shown the figure as reproduced by Stefanescu et al. [1] with dotted and solid lines associated respectively to equation (2) and equation (3). In (**b**) has been drawn the same figure but with the single curve that should have been considered when discussing our calculation results.

Funding: This research received no external funding

Conflicts of Interest: Page: 3

The authors declare no conflict of interest

References

- 1. Stefanescu, D.M.; Alonso, G.; Suarez, R. Recent Developments in Understanding Nucleation and Crystallization of Spheroidal Graphite in Iron-Carbon-Silicon Alloys. *Metals* **2020**, *10*, 221.
- Theuwissen, K.; Lacaze, J.; Laffont, L. Structure of graphite precipitates in cast iron. *Carbon* 2016, 96, 1120– 1128.
- 3. Amini, S.; Abbaschian, R. Nucleation and growth kinetics of graphene layers from a molten phase. *Carbon* **2013**, *51*, 110–123.
- Lacaze, J.; Bourdie, J.; Castro-Roman, M.J. A 2-D nucleation-growth model of spheroidal graphite. Acta Materialia 2017, 134, 230–235.
- Hillig, W.B. A derivation of classical two-dimensional nucleation kinetics and the associated crystal growth laws. *Acta Metall.* 1966, 14, 1868–1869.
- 6. Cahn, J.W.; Hillig, W.B.; Sears, G.W. The molecular mechanism of solidification. *Acta Metall.* **1964**, *12*, 1421–1439.
- Bosze, W.P.; Trivedi, R. On the kinetic expression for the growth of precipitate plates. *Metall. Trans.* 1974, 5, 511–512.
- 8. Hillert, M. Some theoretical considerations in nucleation and growth during solidification of graphitic and white cast irons. In *Recent Research on Cast Iron*; Gordon and Breach: London, UK, 1968; pp. 101–128.
- 9. Geilenberg, H.; Lange, B. Growth experiments with graphite seed crystals from iron melts saturated with carbon. In *The Metallurgy of Cast Iron*; Georgi Pub.: St. Saphorin, Switzerland, 1975; pp. 529–544.
- Ghergu, R.; Magnusson Åberg, L.; Lacaze, J. A possible mechanism for the formation of exploded graphite in nodular cast irons. *Mater. Sci. Forum* 2014, 790–791, 435–440.



© 2020 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access

article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).