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FURTHER DEVELOPMENT AND VALIDATION OF IDS BY MEANS OF SELECTED EXPERIMENTS

Abstract

Microsegregation and material property calculations are the basis for solidification calculations which are essential to understand the continuous casting process. The program IDS (InterDendritic Solidification) is a thermodynamic-kinetic-empirical tool for solidification, microstructure and material properties of steels, which has been developed at Aalto University in Finland since 1984. The heart of the model is the large thermodynamic, diffusion and microstructure data bank made through own assessment work. Particularly, the thermodynamic database of IDS has been clearly extended during the last years.

The quality of the calculations depends significantly on the underlying thermodynamic data. A cooperation has been carried out with the Montanuniversitaet Leoben to further development and validate the IDS databases. A special research field at the Chair of Ferrous Metallurgy in Leoben are high-temperature experiments to investigate the transformation and casting behaviour in the lab, e.g. to identify peritectic steel grades. A portfolio of different laboratory experiments will be presented to validate the calculation results and to further develop the program IDS. A highlight of this work was the optimisation of the silicon and manganese interaction in Fe-C-Si-Mn system based on systematic DSC measurements.

Keywords

Interdendritic solidification, CALPHAD, microsegregatrion, phase diagram, DTA / DSC, HT-LSCM, dipping test.

1. Introduction & Motivation

The use of microsegregation and solidification calculations is essential to describe many phenomena during solidification which cannot be directly measured or observed during the process. This applies particularly for kinetic effects; e.g. the migration of the liquid/solid interface where the separation and diffusion of alloying elements plays a major role (e.g. the calculation of the solidification progress and final solidification point, formation of hot-crack segregation, microstructure development, formation of non-metallic inclusions and precipitates …). Therefore, great efforts have been made to develop suitable numerical models and thermodynamic and kinetic databases. A program that meets these requirements is IDS from Aalto University, which is presented in chapter 2. This program combines thermodynamic equilibrium calculations with kinetic calculation and a variety of material property calculations.

In addition to the model and software development, the validation with literature data and targeted experiments - as described in chapter 3 - plays a major role. Laboratory experiments with defined test parameters and high reproducibility are essential for the validation and further development of such software products. Due to the use of selected alloy variations (by means of model alloys) whole sections of phase diagrams can be constructed

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and individual influences of alloying elements can be investigated in detail. In previous studies [1, 2] from the Chair of Ferrous Metallurgy - based on selected DSC measurements - it was found, that the quite important silicon and manganese interaction in the Fe-C-Si-Mn system was insufficiently described. As the Fe-C-Si-Mn system is of highest relevance for a variety of steels, especially of dual-phase steels, a full section of a Fe-C-1Si-2Mn (wt.-%) system was investigated by systematic DSC measurements and this new results were implemented into IDS. In addition to the equilibrium phase transformations, austenite grain growth was also experimentally investigated and compared with the IDS calculation.

As the transversal cracking during continuous casting depends strongly on the austenitic grain size the grain growth of three selected Fe-C-1%Si-2%Mn alloys was investigated in-situ by high-temperature laser-scanning-confocal-microscopy (HT-LSCM) trials. Furthermore, real solidification experiments were carried out, so-called "dipping tests", in order to examine the initial solidification behaviour in the laboratory. The combined use of selected alloy variations, DTA/DSC-measurements, HT-LSCM observations and "dipping-tests" are very beneficial tools to further develop and validate software tools like IDS, especially for new demanding steel grads.
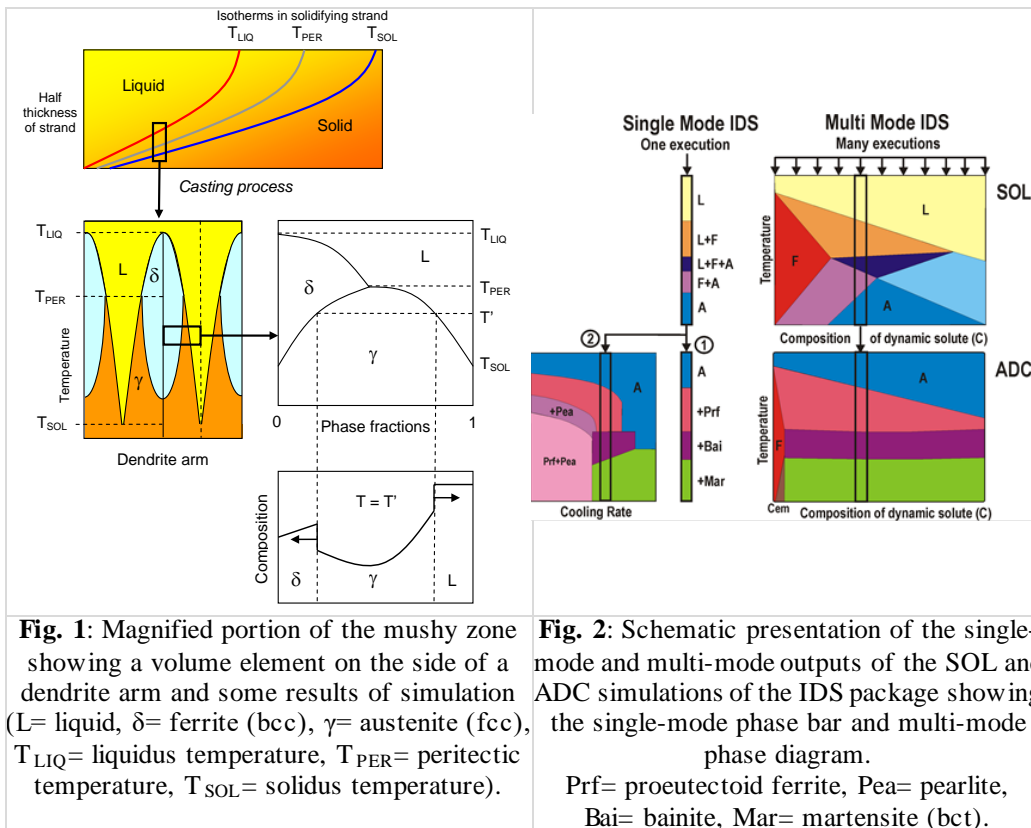
2. Brief description of IDS

The program IDS (InterDendritic Solidification) is a thermodynamic-kinetic-empirical model package for the simulation of interdendritic solidification and microstructure evolution (phase transformations, microsegregation, compound formation, etc.) of steels from liquid to room temperature. The package also simulates solid state phase transformations related to the austenite decomposition process (ADC), at temperatures below 900/600°C, and calculates important thermophysical material properties (enthalpy, thermal conductivity, density, liquid viscosity, etc.). These data are needed in other models, such as heat transfer and thermal stress models, whose reliability heavily depends on the input data itself.

IDS has been developed at Aalto University in Finland and the work has been in progress since 1984. Several papers have been published describing different stages of the ongoing development [3, 4, 5, 6]. Particularly, the thermodynamic database of IDS has been clearly extended during the last years. The calculations of the IDS package are made in one volume element set on the side of a secondary dendrite arm, as visualised in **Fig. 1**. A realistic simulation provides that (1) the dendritic structure forming in different parts of strand is quite regular and (2) there are no strong liquid flows capable of transporting solutes to certain parts of the strand (causing macrosegregation). The present IDS package includes the following calculation modules:

- SOL: simulation of solidification
- ADC: simulation of austenite decomposition
- MAT: calculation of temperature functions for material properties
- GAS: calculation of hydrogen and nitrogen solubility, pressure and diffusivity
- PRF: simulation of precipitate formation at the ferrite/austenite phase interface
- SCA: simulation of oxide scale formation in the strand surface

In detail only the "SOL module" is presented in this work, which can be considered as the main module of the IDS package and is applied in temperature range 1600-900°C. The present solutes available in IDS are - all values are wt-%-max: C (1,5%), Si (2%), Mn (25%), P (0,05%), S (0,05%), Cr (30%), Ni (25%), Mo (6%), Al (2%), Cu (2%), N (0,5%), Nb (0,5%), Ti (0,5%), V (0,5%), Ca (0,02%), B (0,02%) and O, Ce, Mg, H (all 50ppm max).

Fig. 1: Magnified portion of the mushy zone showing a volume element on the side of a dendrite arm and some results of simulation (L= liquid, δ= ferrite (bcc), γ= austenite (fcc), T_{LIQ}= liquidus temperature, T_{PER}= peritectic temperature, T_{SOL}= solidus temperature).

Fig. 2: Schematic presentation of the single-mode and multi-mode outputs of the SOL and ADC simulations of the IDS package showing the single-mode phase bar and multi-mode phase diagram.
Prf= proeutectoid ferrite, Pea= pearlite, Bai= bainite, Mar= martensite (bct).

The SOL module applies thermodynamic chemical potential equality equations (all components), interfacial material balance equations including Fick's 1st diff. law (solutes) and a finite difference application of Fick's 2nd diff. law (solutes). The input data by the user are:
- Nominal composition of the steel
- Cooling rate of solidification (one constant value) or process data of the selected node (time, temperature, deformation).
- Secondary dendrite arm spacing, DAS2 (default value calculated by SOL)

Using these input data the SOL module calculates (for a fixed composition = "single-mode"):
- Solidification of steel (ferritic, austenitic and peritectic solidification)
- Enrichment or impoverishment of solutes in liquid
- Formation of various compounds (inclusions) from the liquid steel
- Homogenization of solute microsegregations below the solidus
- Ferrite-austenite transformation taking place below the solidus
- Formation and dissolution of eutectic ferrite (in high alloy steels only)
- Formation and dissolution of various compounds (precipitates) in the solid structure
- Growth of austenite grains

One new tool is the "Multi-Mode IDS simulation", visualised in **Fig. 2**, where the nominal composition of a selected solute changes by steps. As a result, one gets so called dynamic diagrams illustrating the effect of the varying content of the selected element on special properties of the steel (e.g. a pseudo-binary Fe-C phase diagram).

3. Different laboratory experiments to validate and further develop IDS

A special field of research at the Chair of Ferrous Metallurgy in Leoben is the identification of peritectic steel grades by means of DTA/DSC-measurements in the lab. Beside the investigation of the initial transformation behaviour (pure δ or γ, hypo- or hyper-peritectic) such measurements can be used to determine full phase diagrams. A detailed description of the used DSC setup is published in [7, 8]. The potential of the DSC method is visualized in **Fig. 3** using the Fe-C-Si system as an example.

Three different DSC measurements with different carbon contents from the well-known Fe-C-Si system are compiled. The DSC signal from alloys left of the point C_A (range I = primary δ-Fe solidification) show a clear solidus and liquidus temperature. In addition, alloys of region I show a complete $\gamma \rightarrow \delta$ transformation in the solid before melting.

Alloys between C_A and C_B (range II = hypo-peritectic) exhibit, due to the peritectic phase transformation, a separate characteristic sharp peak which coincides with the solidus temperature. As the peritectic phase transition is a thermodynamic first-order transition which is accompanied by a significant enthalpy step at a constant temperature, this transformation is particularly easy to measure with the DSC. The height of the sharp peritectic peak correlates with the position of the alloy between C_A and C_B. Before the peritectic peak a partial $\gamma \rightarrow \delta$ transformation in the solid takes place.

Alloys between C_B and C_C (range III = hyper-peritectic) exhibit no $\gamma \rightarrow \delta$ transformation in the solid and start to melt in the pure γ region. The DSC measurement clearly shows in the two-phase solid/liquid-region, between T_{Solid} and T_{Liquid}, a separate, sharp peritectic peak.

Apart from these clear criteria for all transformation sequences, all further relevant equilibrium transition temperatures ($T_{\alpha \rightarrow \gamma}$, $T_{\gamma \rightarrow \delta}$, T_{Solid}, T_{Perit}, T_{Liquid}) can also be measured. The investigation from **Fig. 3** clearly shows, that the Fe-C-1wt.-%Si system is very well described in thermodynamic databases (in this case TCFE7 - comparable ones are identical).
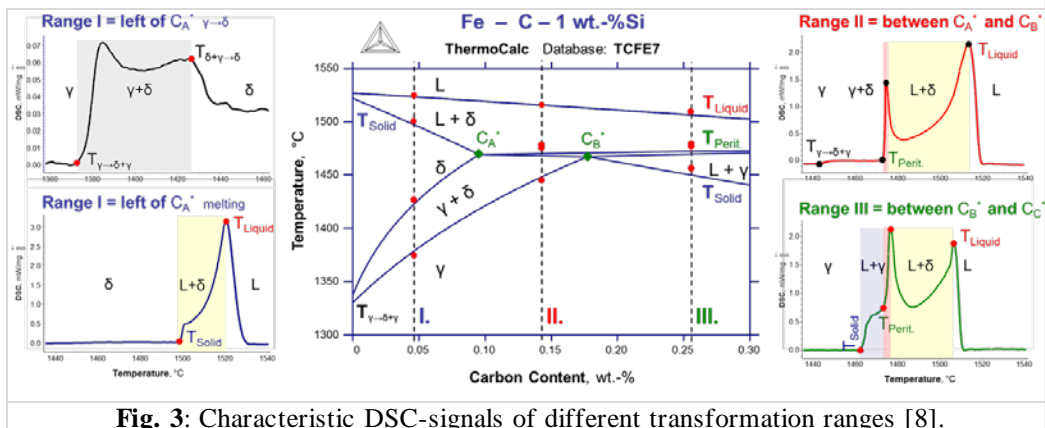

Fig. 3: Characteristic DSC-signals of different transformation ranges [8].

However, such a good match between experimental measurement and thermodynamic calculation is not self-evident. In previous studies - based on selected DSC measurements - it was found, that the quite important silicon and manganese interaction in Fe-C-Si-Mn system was insufficiently described [2]. In order to reassess this important system, small model alloys (about 60g) with different carbon contents of the Fe-C-1%Si-2%Mn (all wt.-%) were produced from high-purity raw-materials in an alumina crucible with a high-frequency re-melting and spin-casting machine under an argon atmosphere, visualised on the right side of **Fig. 4**. Through the use of model alloys the individual influence of alloying elements and their combinations on the Fe-C system can be investigated in detail.

As visualised on the left side of **Fig. 4**, the measured phase transformations are in contrast to the calculated phase diagram (actual SGTE 2014 alloy database). This simple comparison (only 4 alloys) clearly shows that the delta-ferrite range is significantly larger than predicted. A bit higher solidus temperature (T_{Solid}) is predicted, the gamma-to-delta phase transformation ($T_{\gamma \rightarrow \delta + \gamma}$) occurs at much higher carbon content and lower temperatures and the peritectic range occurs at higher carbon content. Finally it should be noted that the peritectic temperature is clearly reduced in comparison to the calculation.
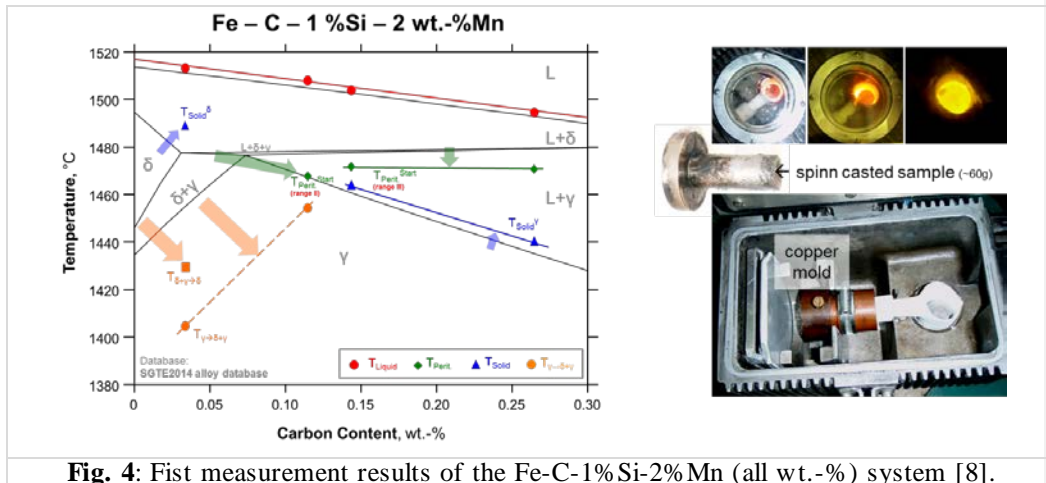
Fig. 4: Fist measurement results of the Fe-C-1%Si-2%Mn (all wt.-%) system [8].
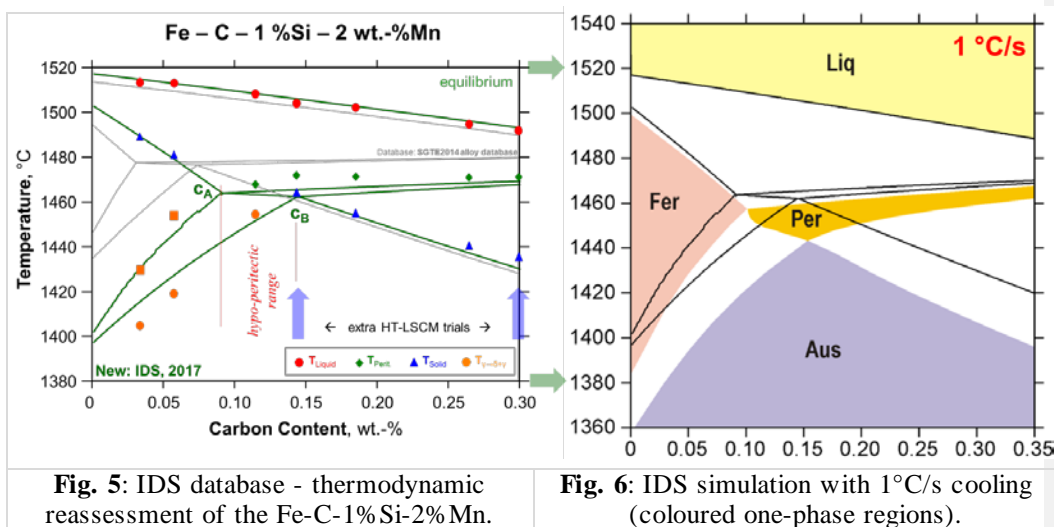

Fig. 5: IDS database - thermodynamic reassessment of the Fe-C-1%Si-2%Mn.	**Fig. 6**: IDS simulation with 1°C/s cooling (coloured one-phase regions).

These new experimental results and some additional points were used for an optimisation of the thermodynamic database of the new IDS (2017). **Fig. 5** shows the optimization results, where a very good agreement between the new measurement points and the IDS equilibrium calculation (Multi-Mode IDS simulation) could already be achieved. In order to achieve this improvement a new quaternary liquid state interaction parameter ($L^{L}_{C,Fe,Mn,Si}$) was implemented. The current investigation demonstrates, that by the use of model alloys (alloy variation) and DSC measurements, phase diagrams and thermodynamic databases can be reassessed.

This new experimentally secured thermodynamic IDS database can now be used for kinetic calculations. **Fig. 5** shows a calculation example of investigated system Fe-C-1%Si-2%Mn (all wt.-%) where the influence of a cooling rate of 1 °C/s is simulated. In comparison to other thermodynamic calculation programs these kinetic calculations of the interdendritic solidification are unique and a great benefit of IDS. Based on such microsegregation simulation, the influence of the cooling rate on the enrichment of alloying elements and the non-equilibrium solidus temperature can be calculated. The knowledge of these results is essential for the subsequent solidification calculation and also for "quality-relevant considerations", e.g. regarding the peritectic solidification behaviour.

In order to validate the peritectic solidification behaviour under initial solidification conditions comparable to the industrial continuous casting process so called "dipping-tests" can be used. Dipping tests are experiments where cold test specimens are immersed into the melt of an opened induction furnace (~20kg melt), as visualised in **Fig. 8**. After a certain time (~10 sec) the test specimens are taken out and after slow cooling under air they are cut into four quarters. This quarter sections (3 dipping tests x 4 quarter = 12 pieces) are scanned and the surface contour are digitized and an automatic evaluation - without human influence - is performed. The calculation of the "unevenness parameter" occurs according the calculation of the "mean slope" of the digitized profile [9]. Former studies from Murikami [10] and Suzuki [11] clearly indicates, that over the carbon content the maximum unevenness index is located between C_A and C_B in the hypo-peritectic range.

Since these tests are much more costly than the fast production of small model alloys and the performance of DSC measurements, only three carbon variations of the Fe-C-1%Si-2%Mn were examined. The diagram in **Fig. 8** clearly shows that the maximum "unevenness parameter" and also the highest standard deviation is also located between C_A and C_B in the hypo-peritectic range (C=0,11 wt-%). This result of a lab solidification experiments stands in excellent conformity with the thermodynamic considerations of **Fig. 4**, **5** and **6**. One big benefit of this "triple-dipping-test" is the high statistical certainty due to the evaluation of the surface contour of 12 pieces.
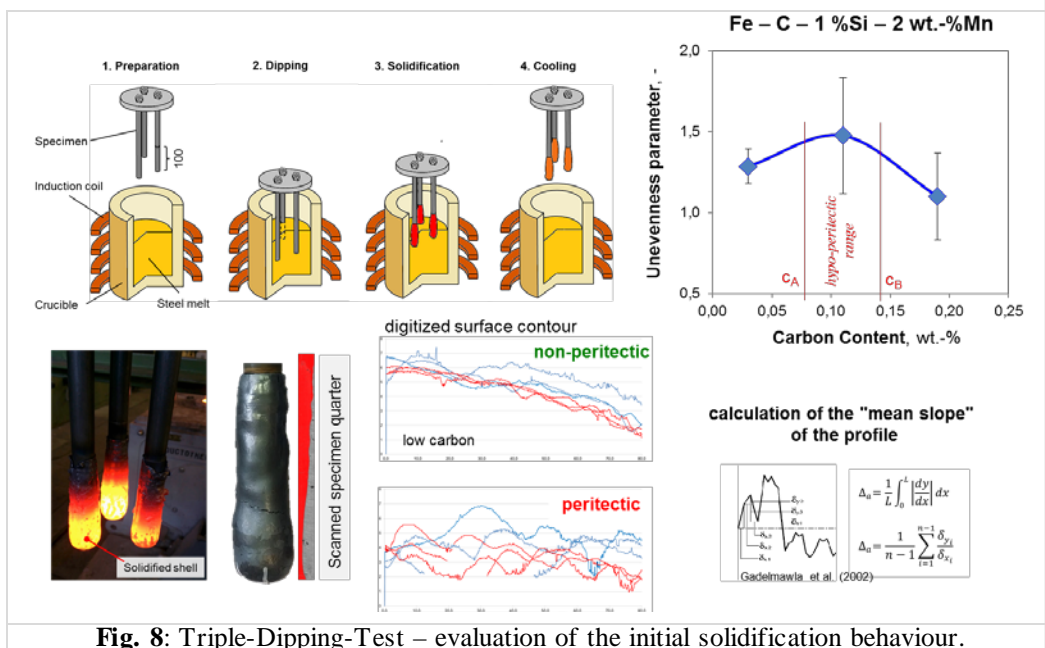

Fig. 8: Triple-Dipping-Test – evaluation of the initial solidification behaviour.

A further field or research is the austenite grain growth and the austenite grain size, which plays an important role in steel production and processing (casting, hot forming, heat treatment), as precipitation and phase transformation kinetics depend strongly on the number of grain boundaries per unit volume (grain boundary density). In plain carbon steels, the austenite grains start to grow either after the completion of delta ferrite to austenite transformation or in case of hypo-peritectic and high carbon steels immediately after solidification [12]. The highest starting temperature and thus the strongest grain growth occur in steel grades with an alloying position at the point C_B (see **Fig. 3**, **5**) where the austenite region has its maximum stability.

Methods for the experimental investigation of austenite grain growth can be distinguished in direct and indirect methods. Indirect measurements are commonly based on the experimental simulation of a thermal cycle and the subsequent metallographic examination of the post-processing sample at room temperature. Direct methods allow the in-situ observation of the microstructure at the respective temperatures. This is possible by means of High-Temperature Laser-Scanning-Confocal-Microscopy (HT-LSCM) which is a special kind of thermo-optical analysis and enables in-situ observations of surface phenomena up to a maximum temperature of 1700 °C. A comprehensive overview on the possibilities of HT-LSCM investigations is given by Sridhar [13].

The HT-LSCM used in this study consisted of a VL2000DX laser scanning confocal microscope, produced by Lasertec, and an attached SVF17-SP high-temperature furnace, and the associated hardware and software from Yonekura. **Fig. 9** gives an overview about the principle design of the HT-LSCM setup, the infrared heating furnace and the sample holder. By using a violet laser beam, whose wavelength is out of the spectra of the sample radiation at high temperatures, it is possible to get high quality images of the microstructure with a resolution below 1 µm (corresponding to a 400 times magnification).
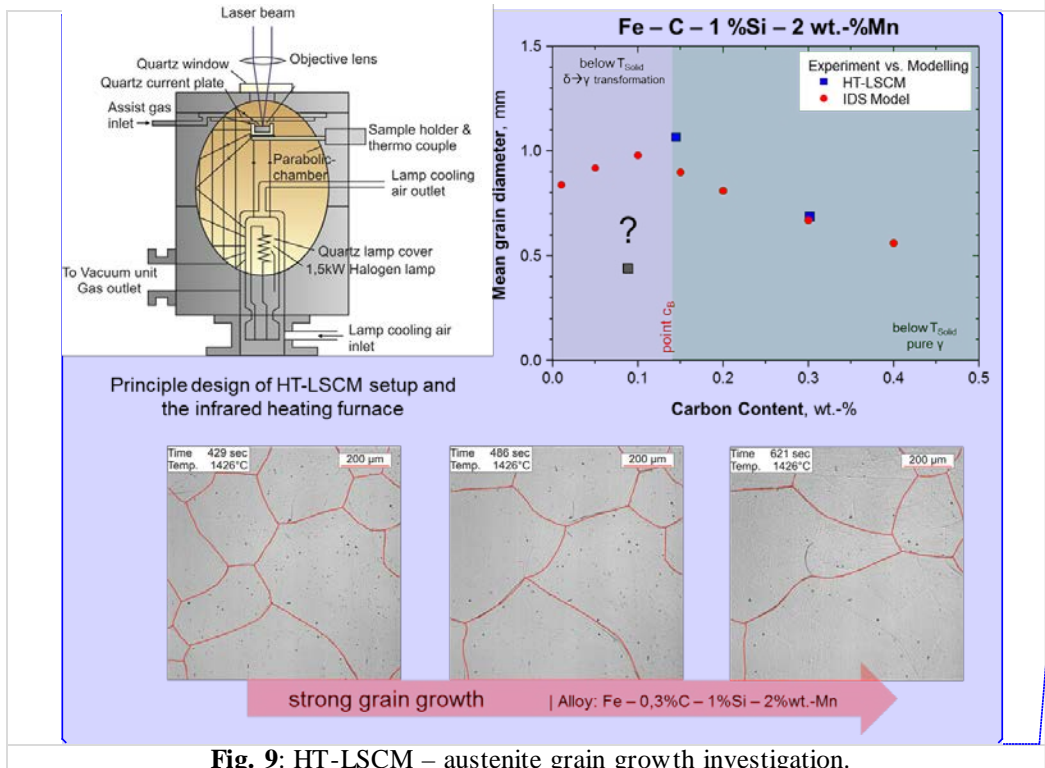
Fig. 9: HT-LSCM – austenite grain growth investigation.

A first investigation of the austenite grain growth was performed with three different carbon contents of the Fe-C-1%Si-2%Mn (same samples as used for the DSC measurements). As this kind of HT-LSCM observation requires a polished surface of the sample, a melting of the sample is not allowed, since the sample would form immediately a droplet which cannot be observed due to the round surface. The prepared samples (polished but not etched) were heated very fast (+500 K/min), kept and annealed 10°C below the solidus temperature to get comparable conditions as "as-cast" and then cooled down (-400 K/min). An in-situ observation of the austenite grain growth is visualised in **Fig. 9** where the movement and dissolution of grain boundaries can be clearly observed. Additional the samples were also subsequent metallographically investigated to get a better overview of the sample due to the larger area that can be evaluated. Nevertheless, the situation with a cold sample which is heated up, but not melted, differs from the IDS calculation which starts in the liquid and simulates a cooling down (with -400K/min). The austenite grain size calculation in IDS is strongly affected by: (1) lack of time (high cooling rate), (2) low temperature, (3) formation of ferrite and precipitates.

A comparison of the experimental results and the IDS model is shown in the diagram - mean grain diameter versus carbon content - in **Fig. 9**. The results for the alloys to the right of C_B appear similar, which is a very promising result, especially since the IDS model was validated by means of various general literature data, but not optimised for this system. Additionally the HT-LSCM clearly confirms that the maximum grain size occurs at point C_B and with higher carbon contents the austenite grain size decreases, similar to T_{Solid}, as visualised in **Fig. 5** – which is an expected result. In contrast, at lower carbon concentrations there is a higher mismatch between HT-LSCM observation and the IDS calculation, In this case, both methods has to be questioned (still an ongoing work area). On the one hand, HT-LSCM investigations at low carbon contents are difficult to evaluate due to the weaker visibility of the grain boundaries; on the other hand, the IDS algorithm has still to be questioned regarding the influence of the delta-gamma transformation. As the transversal cracking during continuous casting depends strongly on the austenitic grain size, such in-situ HT-LSCM observations of grain growth seem to be an interesting method.

6. Summary & Outlook

IDS is a thermodynamic-kinetic-empirical tool for solidification, microstructure and material properties of steels and its ongoing development has a history of over 30 years. This ongoing further development is still necessary due to the increasingly demanding steel grades, especially if they contain increasing quantities of both silicon and manganese. A very important finding in a former study at the Chair of Ferrous Metallurgy was, that in the Fe-C-Si-Mn system, which is one of the most industrially important alloy systems for the design of high strength and high ductility steels, the position of the peritectic range should be at higher carbon content and the δ-region should be larger (bcc-phase more stable).

These new experimental results were implemented in the thermodynamic databases of IDS, in order to perform equilibrium and kinetic calculations. The methodology to re-assess thermodynamic databases by means of self-constructed phase diagrams is highly efficient, especially if the alloy production, the sample manufacture, the DSC measurements, the construction of phase diagrams and the database optimization takes place within a close cooperation. The combined use of selected alloy variations, DTA/DSC-measurements, HT-LSCM observations and "dipping-tests" are very beneficial laboratory methods to further develop and validate software tools like IDS, especially for new demanding steel grads – without the risk and cost of industrial trials.

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