Interphase boundary motion elucidated through in-situ high temperature electron back-scatter diffraction
Zijlstra, G.; van Daalen, M.S.B.; Vainchtein, D.I.; Ocelik, V.; de Hosson, J.T.M.

Published in:
Materials & design

DOI:
10.1016/j.matdes.2017.06.068

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2017

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

Copyright
Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license. More information can be found on the University of Groningen website: https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment.

Take-down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): http://www.rug.nl/research/portal. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.
Interphase boundary motion elucidated through in-situ high temperature electron back-scatter diffraction

G. Zijlstra, M.S.B. van Daalen, D.I. Vainchtein, V. Ocelík, J.Th.M. De Hosson *

Department of Applied Physics, Materials Innovation Institute and Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747AG Groningen, The Netherlands

HIGHLIGHTS

• A new analysis of moving interfaces between ferrite and austenite at high temperatures using in-situ EBSD.
• In contrast to the well-known classical Johnson-Mehl-Avrami-Kolmogorov (JMAK) equation it has been demonstrated that the interphase boundaries move in a jerky-type fashion.
• A linear dependence between the average velocity and the driving force turns out to be a far too simple and a nonlinear description is offered.
• The mean interphase boundary velocity did not critically depend on the crystal (mis)orientations.

GRAPHICAL ABSTRACT

In this work we concentrate on the in situ dynamics of interphase boundary motion during transformations. In-situ high temperature electron-back scatter diffraction (HT EBSD) was employed to study the ferrite-austenite-ferrite transformation in low carbon steel. A novel method was designed to derive the velocity of the interphase boundaries from the EBSD phase maps. It is concluded that the motion of the transformation front occurs in a jerky-type motion, i.e. not continuous in time on a microscale, as the Johnson-Mehl-Avrami-Kolmogorov (JMAK) equation would predict on the macro-scale. It is shown that HT EBSD is capable of providing relevant supplementary insight in the ferrite-austenite phase transformations with adequate spatiotemporal resolution, which would remain hidden in volume averaging experimental techniques such as X-Ray diffraction. In particular, ferrite-austenite phase boundary velocities between 1.4 ± 0.3 and 4.0 ± 0.2 nm/s were detected during isochronal heating with 0.5 °C/min. The mean interphase boundary velocity was ranging between 5.0 ± 0.2 and 6.4 ± 0.2 nm/s for austenite-ferrite transformation with a cooling rate of 1 °C/min. Ledge growth at isothermal conditions, resulted in velocity of 23 nm/s along phase boundary. A strong dependency of interphase boundary mobility and parent-daughter phase boundary misorientation angles was not found.

1. Introduction

Many laws in phenomenological materials physics are based on the concept that a constant force or driving force will lead to a response which is constant in time. In fact, the entire framework of solid mechanics, founded by Isaac Newton himself, is based on this principle. For
example, a constant force per unit of area will lead to a particular deformation response everywhere and at any time in the material; also, a constant driving force will lead to a certain velocity at each point of an interface and to recrystallization and transformation kinetics well described by various phenomenological models. In this paper we concentrate on the dynamics of phase transformations in time as a function of temperature. The Russian mathematician Kolmogorov and American metallurgists Johnson, Mehli and Avrami independently formulated the first mathematical descriptions of isothermal solid-state phase transformation kinetics [1]:

$$f = 1 - \exp(-kt^n),$$  \hspace{1cm} (1)

where \( k \) and \( n \) are *time-independent* constants for a particular reaction. Note that \( k \) cannot be considered a frequency factor as its unit depends on the value of the exponent \( n \). In fact it would be better to formulate the exponent as \((kt)^n\) with \( k \) is per unit of s \(^{-1}\). Nevertheless, this modification will not affect the analysis as Eq. (1) is developed for a constant value of \( n \). The resulting model is nowadays known as the Johnson-Mehl-Avrami-Kolmogorov (JMAK) equation.

More important is to realize that those three essentially equivalent models predict the volume fraction \( f \) converted as a function of time \( t \) in a very large specimen, i.e. boundary effects are ignored and it follows that crystallites nucleate homogeneously throughout the bulk and grow at a constant rate under interfacial control. These models represent the kinetics of a solid state phase transformation as a process of nucleation and growth of a product phase at the expense of the parent phase. The original approach is only valid for random homogeneous nucleation and isotropic growth, but in a modified form it was shown to be applicable for continuous cooling [2] and heterogeneous nucleation [3]. The most important concept in these latter models is that of extended volume, i.e. the volume of new phase that would have been formed if there is no interference from other nuclei.

Nevertheless, this concept has been used very successfully but it breaks down under critical examination, for non-diffusional transformation or under thin film conditions [1,2]. Also, possibly interphase boundaries do not move continuously in time but in a jerky-type fashion, i.e. not in accordance with Eq. (1) when downsizing the observations to a smaller scale. It should be pointed out that the experimental conditions required to observe jerky-type motion are utmost demanding since criteria of high spatial and time resolution have to be met. It is therefore not surprising that the details of the jerky nature of the transformation have gone unnoticed for a long time. With the novel developments in instrumentation it is relevant to revisit the JMAK equation and ask the question whether and when it will break down and at which length scales? Recent evolution of this setup on the speed of in-situ signal processing, allows us to study the dynamics of phase changes with an enhanced temporal resolution. In our work we combine a heating stage and electron-back scattering diffraction (EBSD), resulting in situ scanning electron microscopy (SEM) observations of microstructural changes [4–8].

To test the hypothesis that JMAK will break down we concentrated on ferrite-austenite phase transformations in steel. New generations such as Dual-Phase, TRIP and TWIP achieve mechanical properties from particular mixing of e.g. austenite and ferrite. Therefore, theoretical tools to investigate and predict the transformation interfaces have become increasingly important and with increasing complexity the need to validate the kinetics increases. In addition, in-situ observations of dynamics provide necessary physical insight into the nature of transformations. In most of the theoretical approaches an interface is reduced to a mathematical surface, characterized with variables and parameters as interface thickness, trans-interface diffusivity and interface binding energy [9]. The speed of interphase boundary movement in steel has been reviewed by several authors with experimental techniques as optical microscopy [10], confocal laser microscopy [11] and dilatometry [12,13] (see Table 4). As each technique has its own unique advantage with respect to imaging speed, field of view or sampling volume, information of the crystal orientation cannot be retrieved simultaneously. The uniqueness and novelty of our work lie in making use of the strength of EBSD to accurately determine the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. This allows in-situ observation of nucleation and phase front movement, followed by reverse transformation, i.e. from ferrite to austenite and back to ferrite. For all the microscopy techniques listed above, including EBSD, the derived phase boundary velocity is that of the free surface of the sample; and limited to the usually 2D projection of the (3D) phase boundary.

In particular, we concentrate on the nucleation of a new phase and the mobility of the interphase boundary during transformation. The experimental method to perform EBSD at high temperatures is presented, together with a novel analytical technique to quantify interphase boundary movement.

2. Experimental

The material selected for this in-situ investigation is steel of grade S355, with specifications of chemical composition: maximum 0.24 wt% C and a maximum of 1.6 wt% Mn. Energy-dispersive X-ray spectroscopy (EDS) measurements showed a composition containing Carbon, 0.9 wt% Mn and 0.4 wt% Si. The starting microstructure is ferrite, and the onset of transformation to austenite can be expected at 750 ± 10 °C (\( A_c \)), and should be completed around 911 ± 10 °C (\( A_f \)) [14].

In-situ EBSD experiments were conducted in a Tescan Lyra FEG/FIB Dual beam microscope, equipped with an OIM system by EDAX including a Hikari super camera, which can achieve a maximum of 1400 indexed points per second. Annealing inside the microscope was performed with a Kammrath & Weiss heating module (see Fig. 1), which contains a ceramic resistance heater. As heating is applied from the bottom side, a sample thickness of 0.5 mm was chosen to keep the thermal gradient in the sample to a minimum. The temperature control of the module is calibrated for optimal performance at high temperatures with a maximum of 1500 °C. The temperature could be maintained within a fluctuation of 0.1 °C through a coupled PID-temperature controller. The temperature was monitored through thermocouples at the heating element and just above the specimen.

It is noteworthy that in-situ high temperature EBSD data collection differs from standard EBSD observations in a couple of issues. Although the Kammrath & Weiss heating module is actively water cooled there is a strong thermal radiation from the sample surface directly facing the EBSD phosphorous screen detector. This creates an additional back-ground signal, which has to be subtracted from the collected Kikuchi patterns. At constant temperature the background could be collected just once for both: non-diffracted back scattered electrons and thermal radiation component. However, when temperature is changed the gain parameter for the CCD camera recording the EBSD signal has to be tuned for optimal Kikuchi pattern recognition. This procedure requires an additional time to optimize observations at different temperatures. Another issue is the limited cooling ability of EBSD detector itself, which may lead to the substantial temperature increase and even to thermal damage of phosphorous screen, especially for sample temperatures over 750 °C. We placed another thermocouple at its vicinity and retracted the whole EBSD detector for a while when its temperature exceeded 120 °C. Data collection is therefore limited to times < 100 s at temperatures higher than 800 °C. Finally, a problem with the drift of image, appearing mainly at moments when a substantial change in heating rate is required, has to be minimized/corrected. We did not perform OIM scans at these conditions and final drift was always corrected by placing a selected object (e.g. small carbide particle or surface feature) into the SEM image center, which could be performed very quickly. Measurements with the forward-scattered-electron detector were not performed, since this detector has been removed to avoid damage to it at high temperatures.
EBSD data collection was done with the use of the TSL OIM Data Collection 7.3 software. OIM analysis was performed by TSL OIM Analysis v.7.3 software. This included a two step data cleaning procedure with Grain Confidence Index Standardization (grain tolerance angle 5°, minimum grain size of 5 pixels, with a condition that grain contains multiple rows) and a Neighbor Orientation Correlation (level 4, tolerance 5, minimal Confidence Index 0.1). All remaining data points with a confidence index below 0.1 were removed and are shown as white points in OIM maps. It is worthwhile to note, that always index below 0.1 were removed and are shown as white points in OIM maps. In Fig. 2, where the dashed line represents the average interphase boundary position. The operation was repeated for each successive map, whereby all new lines were drawn parallel to the initial phase boundary position. The operation was repeated for each successive map, resulting in an average speed of 4.0 ± 0.2 nm/s. Then it made a sudden jump between two scans, as Fig. 4.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The time to make a mapping with EBSD is initially prescribed by the size of the map and the density of scanning points. Secondly, the hypothesetical processing speed may suffer if the indexation time per Kikuchi pattern increases due to decreasing signal quality. This can require multiple frames averaging to improve the signal to noise ratio. Observation of the dynamics of interphase boundary movement demands rapid successive mapping. Therefore, selection of scanning parameters is a trade-off between map size and spatial resolution (step size). This results in typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The time to make a mapping with EBSD is initially prescribed by the size of the map and the density of scanning points. Secondly, the hypothesetical processing speed may suffer if the indexation time per Kikuchi pattern increases due to decreasing signal quality. This can require multiple frames averaging to improve the signal to noise ratio. Observation of the dynamics of interphase boundary movement demands rapid successive mapping. Therefore, selection of scanning parameters is a trade-off between map size and spatial resolution (step size). This results in typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.

The strength of EBSD is the accurate determination of the phase front within sub-micron error, combined with the measured crystal orientation of the parent and daughter grains. In this work a novel analytical method was developed to derive the velocity of the interphase boundary from the EBSD phase maps. In this method partly transparent typical map parameters of a 30 × 30 μm² scanning area with step size 0.4 μm in a hexagonal type of grid, and an indexing speed of 50 points per second. The time between successive scans was 85 s, with an additional 15 s in isochronal experiments, when in the latter case background collection and correction had to be applied due to its change with temperature.
b) shows. It is possible to give an estimate of the lower limit of this jerky motion jump of 40–90 nm/s.

For the determination of the velocity of the interphase boundary, the position of the boundary itself was averaged. As illustrated in Fig. 2, the boundary is not straight. When the interphase boundary shifts, it does not maintain a similar waviness nor the same amount of kinks. Fig. 6 shows the evolution of the interphase boundary shape. Clearly rough and wavy fronts as in a) c) j) can be discriminated from relative straight ones like e) and g).

Besides the crystal orientation, interphase boundary movement and waviness, also the phase fractions as a function of time can be determined from the EBSD maps. The increase of the fraction of austenite during heating with 0.5 and 1.5 °C/min, is shown in Fig. 7.

Using the general mathematical form of the JMAK equation of Eq. (1) the reaction rate indicated by factor n can be derived by the determination of the slope of ln [−ln (1 − f(t))] vs. ln (t). A value of n = 4.8 ± 0.1 was found for heating with 1.5 °C/min. However, two stages of growth with different speeds were observed when heating with 0.5 °C/min: first stage n = 1.39 ± 0.03, and second stage n = 4.5 ± 0.1. Below 840 °C the main contribution to the increase of the austenite fraction is due to the fact that an existing austenite grain extends along the grain boundary. The jump in fraction of austenite at 840 °C (time = 35 min) corresponds to the rapid interphase boundary movement of the grain with 48° misorientation at the top left of Fig. 4. This event also marks the onset of the second stage of transformation with higher reaction rate. The contribution to the fraction of this second stage is a summation of both steady growth and boundary jumps. Steady growth and jumps were also observed for heating with 1.5 °C/min, however this did not result in two stages of growth in the austenite fraction. The higher reaction rate and the limited temporal resolution, already disables to visualize the individual contribution of the two processes.

In another experiment local reverse transformation was observed. Here, the sample was heated to 800 °C with 10 °C/min, followed by heating to 820 °C with 2 °C/min. Austenite grains were already visible on the first scan at 700 °C, and at 750 °C three more austenite grains had nucleated. A large amount of ferrite grain recrystallization was observed at 814 °C, as shown in Fig. 8. Instead of new austenite grain nucleation, the size of two of the austenite grains had decreased and one of these grains disappeared. The driving force for recrystallization seems to have increased during the heating. The reverse transformation can be assisted by a change in the carbon gradient. Carbon diffusion from the phase boundary into the austenite grain can cause a local carbon depletion zone. The drop in carbon concentration in this binary interphase temperature range, makes the ferrite phase temporally more favorable.

In the above experiments, austenite nucleation temperatures were found of 895 °C, 881 °C, 840 °C and below 700 °C. It can be concluded that surface transformation deviates from the expected bulk
transformation temperatures. This scatter in starting temperatures is probably influenced by the history of the material such as heating ramp and holding time at certain temperatures, the presence of a high-energy interphase boundary at the extreme surface, as well as small differences in sample thermocouple location.

3.3. Velocity of interphase front during cooling

A fully austenitic sample was cooled from 860 °C with a cooling rate of 1 °C/min. The first ferrite was observed at 802 °C. The decreasing fraction of austenite during cooling also shows JMAK behavior. The absolute value of \( n = -18.7 \pm 0.6 \) is much larger compared to the found value of \( n = 4.8 \pm 0.1 \) for heating with 1.5 °C/min. Although the temperature ramp is smaller, the back transformation from austenite to ferrite is much faster. Nucleation of the ferrite grains occurred outside the scan area. The first two incoming grains moved along the austenite grain boundary, similar as interphase growth during heating. At 783 °C a ferritic grain is penetrating the center of an austenitic grain, as shown in Fig. 9, for which jerky motion was observed. The mean velocity was ranging between 5.0 ± 0.2 and 6.4 ± 0.2 nm/s (Table 2). The precise dynamics of this accelerated stage could not be observed due to limited number of scans. Estimation of the lower limit of the speed yields about 130 nm/s. It is concluded that jerky motion is thus also observed during cooling.

3.4. Velocity of interphase front at constant temperature

In the aforementioned experiments the temperature was increased at constant rate (isochronal) in order to reveal the nucleation sites and dynamics of interphase growth. The following isothermal experiment was performed to study these phenomena in quasi-static conditions.

The specimen was heated up and kept at 820 °C for 20 min. Thereafter heating continued to 840 °C with 0.5 K/min. First small changes started to occur at 835 °C. Although a new grain nucleated between 830 °C and 835 °C, very little growth took place in the following 37 min afterwards. Austenitic grain growth became more pronounced at 840 °C. Two new grains nucleated, one after 17 min and one after 25 min. As observed before, expansion of the austenite phase took place mostly along the grain boundaries. At this temperature a ferrite grain became surrounded by austenite grains. As shown in Fig. 10, the austenite grains grew radial inwards with small steps. Often a step or ledge was formed on the interphase boundary, followed by boundary growth along this ledge. Fig. 10(b)–d) shows this growth, for which a ledge velocity of 23 nm/s can be derived. On a few occasions also reverse transformation took place along the same ledge, to be succeeded again by ferrite-austenite transformation. The mean velocity of the inwards moving interphase boundary ranged between 0.4 ± 0.2 and 1.7 ± 0.3 nm/s, as displayed in Table 3. No clear correlation can be seen between the average velocity and the misorientation between the parent-daughter interphase boundaries. It was observed that twin daughter fractions grew slightly faster.

A new grain nucleated on a grain-boundary while heating to 845 °C. This grain had the K\(^{-}\)S OR with the ferrite grain on one side of the nucleus, but grew mostly in the direction opposite to this OR in the ferrite grain on the opposite side which did not have the K-S OR. As expected the incoherent interphase boundary which does not have the K-S OR,

![Fig. 4](image_url) Heating from 820 °C with 1.5 °C/min. Overlaid phase maps for a) early stage of transformation at around 835 °C, and b) nearly full transformed around 847 °C. Ferrite displayed in red and austenite grains in green. Growth directions are indicated by the black arrows, together with the grain misorientation angles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

![Fig. 5](image_url) Interphase boundary motion during heating with 0.5 °C/min for several interphase boundary angles.

### Table 1

<table>
<thead>
<tr>
<th>Misorientation (°) (±0.5)</th>
<th>Average speed (nm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>48</td>
<td>4.0 ± 0.2</td>
</tr>
<tr>
<td>46a</td>
<td>1.5 ± 0.2</td>
</tr>
<tr>
<td>46b</td>
<td>3.3 ± 0.5</td>
</tr>
<tr>
<td>42</td>
<td>2.5 ± 0.5</td>
</tr>
<tr>
<td>41</td>
<td>1.4 ± 0.3</td>
</tr>
</tbody>
</table>
provides a greater driving force to move this interphase boundary at a higher mean velocity, than the semi-coherent K-S OR boundary.

4. Discussion

We performed a careful literature search so as to place our findings in the right context. For decarburization experiments, with a macroscopic moving interphase front, an interface velocity between 0.01 and 0.1 μm/s is reported [19] (but note: from austenite to ferrite). The velocities of the planar front that are ranged between sub-micrometer to nanometer per second [9]. It is shown [20] that ferrite growth under decarburization following parabolic growth kinetics. Schmidt et al. [21] used heating rates of 3 to 10 °C/s and found that the migrating austenite front in interstitial free steel covers a distance of about 200 μm in 1 s. An overview is listed in Table 4. As the listed techniques are limited to observations in one plane, the velocities presented are 2D projections of the (3D) phase boundary, which is similar for the phase boundary velocity derived with EBSD.

Overall the interphase boundary velocities in austenite-ferrite transformation are a little higher compared to the ferrite-austenite transformation, but still in the same order of magnitude. It is difficult to say to what degree the velocities derived in this work can be compared with respect to each other. The difference, however, is believed to be the result of the higher cooling ramp (1 °C/min) as compared to the heating ramp (0.5 °C/min) and undercooling. Velocities for the austenite-ferrite transformation in pure Fe and ultralow carbon steel have been reported (Table 4), with differences in heating rates, 1 °C/min used here opposed to 5–20 °C/min in literature, show velocities differing by a factor 1000, i.e. nm/s in this work vs. μm/s in literature. Post-mortem analysis of isothermal decarburization of Fe-C-Si steel, provided derived speeds of 0.1–0.01 μm/s [19]. Low impurity content, together with high cooling speeds seems to result in phase boundary speeds in the order of at least μm/s. The discrepancy is probably not only due to the difference in heating rate, but also due to the decrease in solubility of carbon.

In a few rare cases growth speeds have been presented in literature for phase transformations in pure iron and steel. From dilatometry data an average speed of 3.05 μm/s was found in the case of the austenite-ferrite transformation in pure Fe for cooling rates of 5–15 °C/min [12]. In similar experiments an average speed of 1.6 μm/s was found for the austenite-ferrite interface of a Fe-0.01 at.% C at 20 °C/min cooling, as compared to 3.9 μm/s for pure iron at the same cooling rate [13]. In the same paper a transition from diffusion-controlled growth to interface-controlled growth was discussed. Diffusion-controlled growth was attributed to the initial stages of transformations for cooling rates of 5–10 °C/min. Velocities of about 5 times smaller (~10⁻⁷ m/s) than for massive interface-controlled growth, were derived from the average grain size diameter of small grains.

In optical microscopy experiments the ferrite-austenite transformation of Fe-4.2 at.% Cr with 5 °C/min heating showed two stage transformation behavior with 0.33–0.75 μm/s and a stage with 3.7–7.6 μm/s, respectively [10,22]. The two stage behavior was believed to be due to a change in driving force related to the phase transformation from the ferrite-austenite coexisting region to the austenite single phase region of the Fe–Cr alloy. This two stage behavior was also observed for heating at a rate of 0.5 °C/min. In the first stage the average velocity was 4.0 ± 0.2 nm/s, followed by a jump of 40–90 nm/s. The two stages presented in this work differ by an order of magnitude, similar as observed in [22]. It is questionable whether the temperature of 840 °C is the actual transformation temperature for fully austenite (in [22] attributed as the driving force), as other grains made a jump later, or not at all. Evidence for pinning, due to an oxide layer or precipitates as the origin of jerky motion was not found: precipitates were not observed, and an oxide layer is less likely either since it could block the EBSD signal.

The existence of both boundary morphologies, i.e. ragged and straight, indicates a jerky-type interphase boundary movement mechanism, of a smaller order than the resolution reported in Figs. 5 and 6. In fact all interphase boundaries in our case seem to move in a jerky manner, i.e. observations reveal a segmental motion of the interphase boundary, where segment length, stagnation period and local velocity may vary. By this mechanism, the boundary roughens by small segments of the new phase moving forward. When the boundary has a certain degree of waviness and roughness and therefore an amount of stored energy, a "jump" sideways reduces the boundary length.

We ask ourselves: What determines the characteristic time scale and what affects the characteristic length scale? As aforementioned it should be pointed out that the experimental conditions required to observe jerky-type motion are most demanding since criteria of high spatial and time resolution have to be met simultaneously. In the present experimental set-up we are limited in spatial and temporal resolution.
and to put the in-situ EBSD-SEM observation in perspective let us define the spatio-temporal resolution $\delta_{ST}$ as the product of spatial and resolution in time:

$$\delta_{ST} = \delta_S \otimes \delta_T.$$  \hfill (2)

To make an estimate of the spatiotemporal resolution based on FEG-Lyra Tescan in SEM mode (not the same as EBSD mode) the spatial resolution is about 4–6 nm when using accelerating voltages ranging between 15 and 30 kV. Although we have employed a Hikari super camera which can achieve a maximum of 1400 indexed points per second, the actual EBSD processing speed may still suffer if the indexation time per spot increases due to decreasing signal quality. A conservative number is indexation of 50 spots per second, with a typical step size of 0.4 $\mu$m, this results in $\delta_{ST} = 8.0 \times 10^{-9}$ m⋅s. However to construct a $30 \times 30$ $\mu$m map (one frame) with these parameters requires indexation of 6380 points, which leads to $\delta_{ST} = 5.1 \times 10^{-5}$ m/s. The time between successive scans was around 85 s. In comparison, in-situ TEM experiments (not EBSD but dark field/bright field observations) to date offer a time resolution of about 30 frames per second with a spatial resolution of sub-nm leading to $\delta_{ST} = 10^{-11}$ m/s (at best).

As shown in Section 3, the in-situ observations at low heating rate of 0.5 °C/min reveal a splitting in transformation reaction rates. First a slow growth along grain boundaries and inward ferrite parent grains with $n = 1.39 \pm 0.03$, followed by large steps of the interphase boundaries resulting in $n = 4.5 \pm 0.1$. It can be concluded that the motion is far from continuous, neither on a macroscale as analyzed with the help of JMAK Eq. (1) or on a local scale as observed in our experiments (see Section 3). The results of Fig. 7 not only indicate that the heating rate is a process-determining factor here, but also demonstrate that in-situ HT EBSD is capable of providing insight in:

- trends at both micro- and macroscopic scales: even on a small area of $40 \times 40$ micron squared clear stages of nucleation, growth and saturation can be observed, resulting in an austenite fraction curve with a typical JMAK shape.
- local phenomena like two different transformation phenomena in time, which would remain hidden in more volume averaging experiments, e.g. by using X-ray diffraction.

The competition between continuous motion and jerky type motion is determined by the driving force. The argument, as has been pointed out by John Cahn [23,24] is that jerky type motion will take place below a certain critical driving force whilst above a sufficiently large force field continuous motion will always be possible. For an infinitely flat interface the driving force to move the entire interface will not be easily achieved in practice, whereas in case of a highly thermodynamically diffuse interface, i.e. with a lot of a preexisting disorder along the interface plane, any driving force will make it moving in a jerky fashion. In the present case of heterophase interfaces as observed in the present

<table>
<thead>
<tr>
<th>Misorientation (°) (±0.5)</th>
<th>Average speed (nm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>38</td>
<td>6.4 ± 0.2</td>
</tr>
<tr>
<td>37</td>
<td>5.0 ± 0.2</td>
</tr>
</tbody>
</table>

**Table 2:** Average speed of the interphase boundary during cooling with 1 °C/min, for several parent-daughter interphase boundary angles.
where $\phi$ is equal to unity for an infinitely straight front and decreases to lower values for ragged configurations. Substituting the appropriate values for $\Delta g_{\alpha\gamma}/\Omega$ of $0.2 \times 10^7$ J/m$^3$ at $840$ °C, $\Gamma_{\alpha\gamma}$of $600$ mlj/m$^2$ [10] and $\nu$, of the order of sub-nanometer into Eq. (3) leads to the conclusion that a continuous motion is not expected in the present case, i.e. the transformation front advances not continuously but rather by the motion of steps, kinks, ledges for the case of Kurdjumov-Sachs (111)fcc||(110)bcc orientation relationships along the interphase boundary and roughening for transformations along grain-boundaries.

The physical picture which emerges from our experiments and these theoretical considerations is demonstrated in Fig. 10. Obviously the mean velocity will depend on the correlation length along the transformation front. Further the mean displacement of the front depends on the local correlation length, i.e. the local step height or roughness amplitude, say $\xi$, whereas the time involved depends on the range of interaction, say $\xi'$. Since there will be only a physical correlation length when the jerky type regime is operational, i.e. below a critical driving force $F_c$ above which jerkiness becomes unlikely, the correlation length scales, taking into account Eq. (3), with:

$$\xi = \left( \frac{\Delta g_{\alpha\gamma}/\Omega}{\Gamma_{\alpha\gamma}} \right)^{\frac{1}{\mu}} \left( \frac{\Delta g_{\alpha\gamma}/\Omega}{\nu} \right)^{\frac{1}{\nu}} \xi'$$

Table 3

<table>
<thead>
<tr>
<th>Misorientation (°) ±0.5</th>
<th>Average speed (nm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>54</td>
<td>0.4 ± 0.2</td>
</tr>
<tr>
<td>53</td>
<td>1.7 ± 0.2</td>
</tr>
<tr>
<td>40</td>
<td>1.5 ± 0.2</td>
</tr>
<tr>
<td>48</td>
<td>1.4 ± 0.3</td>
</tr>
</tbody>
</table>

Table 4

<table>
<thead>
<tr>
<th>Material</th>
<th>Transformation</th>
<th>Temp. ramp (°C/min)</th>
<th>Velocity (μm/s)</th>
<th>Technique</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe-4.2 at.% Cr</td>
<td>Fer.-Aus.</td>
<td>5</td>
<td>0.33–7.6</td>
<td>Optical Confocal laser</td>
</tr>
<tr>
<td>IF steel</td>
<td>Fer.-Aus.</td>
<td>180–600</td>
<td>200</td>
<td>Dilatometer</td>
</tr>
<tr>
<td>Pure Fe</td>
<td>Aus.-Fer.</td>
<td>5–15</td>
<td>3.05</td>
<td>Dilatometer</td>
</tr>
<tr>
<td>Fe-0.01 at.% C</td>
<td>Aus.-Fer.</td>
<td>20</td>
<td>1.6</td>
<td>Dilatometer</td>
</tr>
<tr>
<td>Pure Fe</td>
<td>Aus.-Fer.</td>
<td>20</td>
<td>3.9</td>
<td>Dilatometer</td>
</tr>
<tr>
<td>Fe-0.76C-0.84Si</td>
<td>Aus.-Fer.</td>
<td>Isothermal 0.1–0.01</td>
<td>Post Mortem</td>
<td>[19]</td>
</tr>
</tbody>
</table>

Fig. 10. a) Overlaid [001] inverse pole figure maps indicating interphase boundary growing directions and misorientation at a constant temperature of 840 °C. The large grain in the center is ferritic, and surrounded by new austenite grains. b) Ledge step $\xi'$ equal to scan step size of 0.75 μm, and ledge correlation length $\xi$. c), d), e): Section of the 49° interphase boundary at 840 °C, time between scans is 65 s. Recorded expansion of the interphase boundary ledge along $\xi'$ with two steps of 1.5 μm.
To investigate the scaling exponents this rather formal formulation of Eq. (5) can be confronted to the experimental observations and to the macroscopic mean velocity as predicted by simple thermodynamics. As abovementioned the snapshots of Fig. 6 are random frames of the repetitive process of boundary straightening and kink/ledge roughening. From Fig. 6, g) a line segment of at least 4 μm is straight. Due to the hexagonal scanning pattern with a step size of 0.4 μm, the boundary appears rather ragged. Hence the maximum boundary position uncertainty here is half the step size: 0.2 μm. Fig. 6f) has several kinks, resulting in a boundary width of 1 μm. Although the jump time is unknown due to the limited temporal resolution, it is shown that the maximum observed jump perpendicular to the interphase boundary of the whole segment presented in Fig. 6, is 1 ± 0.2 μm. It must be realized that this phase transformation may not take place in a plane, but rather in three dimensions.

Within a classical approach the mean velocity for the ferrite-austenite transformation upon increasing the temperature can readily be derived from the net transfer of atoms from ferrite to austenite per unit of time (assuming ferrite and austenite all have the same chemical composition which may not be the case because of carbon segregation in carbon-rich and dissolution of carbides at higher T). The mean velocity is written as:

\[
\bar{v} = \lambda v_o \exp \left[ -\frac{\Delta G_a + E_a}{kT} \right] - \lambda v_o \exp \left[ -\frac{\Delta G_a + \Delta G_{gy} + E_f}{kT} \right],
\]

where \(\Delta G_a\) is the activation energy for one atom to leave ferrite, crossing the interface and attach to austenite. The frequency of jumping atoms back and forth across the interface is taken to be identical in ferrite and austenite at the corresponding temperatures. \(E_a\) and \(E_f\) refer to strain energies in gamma-austenite and alpha-ferrite, respectively and \(\Delta G_{gy}\) is the change in free energy for the growth of austenite by the atoms coming from ferrite. For the moment, the mean velocity derived from simple thermodynamics can be written as:

\[
\bar{v} = \lambda v_o \frac{\Delta G_{gy}}{kT} \exp \left[ -\frac{\Delta G_a}{kT} \right].
\]

\(\lambda\) is the distance across the interface and \(v_o\) the characteristic frequency (kT/h in Eyring’s theory). Taking the values from literature the estimated mean velocity is found 1.5 nm/s at 840 °C which is in the right order of magnitude (see Table 3) and maybe a bit fortuitous in accordance with the experimental observations. It is assumed that at the onset of the transformation the concentrations of carbon atoms across the transformation front differ considerably (maximum carbon concentration in austenite is 0.011 and 0.001 in ferrite at 840 °C). However, carbon atoms may have moved over quite a large distance away from the front within the timeframes applied in our experiments. This research was carried out under the project number T63.3.12480 in the framework of the research program of the Materials Innovation Institute M2i, Delft, the Netherlands.

Dr. René de Kloe from Edax and Dr. Volker Stirić from Kammrath & Weiss are acknowledged for their help with the HT EBSD system configuration.

5. Conclusions

We have presented a new analysis of moving interphase boundaries between ferrite and austenite at high temperatures using in-situ EBSD. The results are confronted to the well-known classical Johnson-Mehl-Avrami-Kolmogorov (JMAK) equation. It is observed that the interphase boundaries do not move continuous in time but in a jerky-type fashion. In particular, based on in-situ EBSD observations as a function of temperature we conclude that the nucleation takes place on grain-boundaries and triple junctions and that growth occurs along grain boundaries. A linear dependence between the average velocity and the driving force related to the change in Gibbs free energy between the phases as predicted by a classical thermodynamical approach turns out to be a far too simple and the non-linearity as formulated in Eqs. (5) and (8) offers a better theoretical framework.

References
