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Phase transformation modeling and parameter identification from dilatometric investigations

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Abstract. The goal of this paper is to propose a new approach towards the evaluation of dilatometric results, which are often employed to analyse the phase transformation kinetics in steel, especially in terms of continuous cooling transformation (CCT) diagram. A simple task of dilatometry is deriving the start and end temperatures of the phase transformation. It can yield phase transformation kinetics provided that plenty metallographic investigations are performed, whose analysis is complicated especially in case of several coexisting product phases. The new method is based on the numerical solution of a thermomechanical identification problem. It is expected that the phase transformation kinetics can be derived by this approach with less metallographic tasks. The first results are remarkably promising although further investigations are required for the numerical simulations.

1.   Introduction

Dilatometry is a means to measure material properties which are related to the change in volume. A dilatometer is able to measure this volumetric change in one dimension, which is often related to the change in temperature. Steel dilatometry is of special importance for the investigation of γ/α-phase transformation kinetics [1, 2] as the differ-ent phases exhibit different crystallographic structures. The temperature, where the phase transformation occurs, can be detected precisely by correlating the temperature and the specimen's change of length. The change of length according to temperature is fairly constant in contrast with the one caused by phase transformation. During phase transformation, the specimen expands during cooling although the temperature decreases as a result of the microstructure rearrangement. While the determination of the start and end of a phase transformation is relative simple, the determination of phase transformation kinetics requires additional metallographic investigations, in which the samples are grinded, polished, etched and observed under light optical microscope [3]. By deriving different phase fractions from different cooling rates in the experiment, the entire phase transformation kinetics in terms of continuous cooling transformation (CCT) diagram can be defined. The fractions of product phases are determined from the metallographs by a purely visual judgement of the experimenter or with the aid of image analysis software, which work under a pixel based principle, i.e. different phases are distinguished according to their colours. Nevertheless, in addition to experimenter's experience, the determination of phase fraction depends also on the homogeneity and phase distribution in the sample as the metallographic observed areas are not truly representative for the entire specimen.

In a recent paper [4], it has been shown that at least in case of no more than two coexisting product phases, the information of the complete transformation kinetics is already derivable from the two time-dependent values of length-change and temperature from a single point measurement in the dilatometer. Thus it should be possible to avoid further metallographic investigations in this case.

The purpose of this paper is to validate this finding for the case of general dilatometric data and to develop a strategy to extract as much information as possible from dilatometer experiments.

2.   Mathematical View on Dilatometer Experiments

2.1.   The direct problem

A standard shape of dilatometer specimen is a cylinder with height L. In the sequel, it is assumed that radial and azimuthal variations of the physical quantities: temperature T and displacement u are negligible; $z = (z_1, z_2, z_3, z_4)$ stands for the vector of product phases, i.e., ferrite, pearlite, bainite and martensite.

One of the simplest models to describe thermal expansion as illustrated in **Figure 1** is taken by assuming a mixing rule for thermal strain, i.e.

$$\varepsilon^{th} = \sum_{i=0}^{4} z_i \, \varepsilon_i^{th},$$

where the thermal strain in each phase is given by the linear model $\varepsilon_i^{th} = \delta_i (T - T_i^{ref})$, with a thermal expansion coefficient δ_i and a reference temperature T_i^{ref}. Note that the remaining phase fraction of austenite is given by $z_0 = 1 - z_1 - z_2 - z_3 - z_4$.

For convenience, the following nomenclatures are introduced

$$\alpha_i = \delta_i - \delta_0 \text{ and } \beta_i = \delta_i T_i^{ref} - \delta_0 T_o^{ref} \quad \text{for } i = 1, \ldots, 4$$

as well as

$$\delta(z) = \delta_0 + \sum_{i=1}^{4} \alpha_i z_i, \eta(z) = \delta_0 T_o^{ref} + \sum_{i=1}^{4} \beta_i z_i.$$

Then, the thermal strain is given as

$$\varepsilon^{th} = \delta(z) \, T - \eta(z).$$

Assuming further an additive partitioning of the overall strain into a thermal one and an elastic one, the following quasi-static, linearized thermo-elasticity system is obtained:

$$\left(u_x - \delta(z)T + \eta(z) \right)_x = 0, \tag{1a}$$

$$\rho c T_t - k T_{xx} + \Lambda \delta(z) u_{xt} T - \rho L \cdot z_t = \gamma (T^{ext} - T). \tag{1b}$$

Since cooling happens all around the specimen, the distributed Newton type of cooling law has been chosen with a heat transfer coefficient γ and a coolant temperature T^{ext} ; ρ is the density, c the heat capacity, k is the thermal conductivity and $L = (L_1, \ldots, L_4)$ the vector of latent heats of the product phases. $\Lambda = 2\Lambda_1 + \Lambda_2$ is the bulk modulus with the Lamé constants Λ_1, Λ_2.

The system has to be completed with boundary and initial conditions. It is assumed that the specimen is isolated at its end points $x=0$ and $x=L$, i.e.

$$T_x(0, t) = T_x(L, t) = 0 \tag{1c}$$

Moreover, it is fixed on the left-hand side and stress free on the right-hand side, i.e.

$$u(0,t)=0 \quad \text{and} \quad u_x(L,t) - \delta(z)T(L,t) + \eta(z) = 0. \tag{1d}$$

To compute the phase fractions, one can add the Avrami-type kinetics [5-7] or rate laws as in [8, 9]. Afterwards the direct problem is completely defined and for a given set of boundary and initial conditions, the solution (u,T,z) can be computed. However, in this paper another viewpoint has been oriented and will be explained in the following subsections.
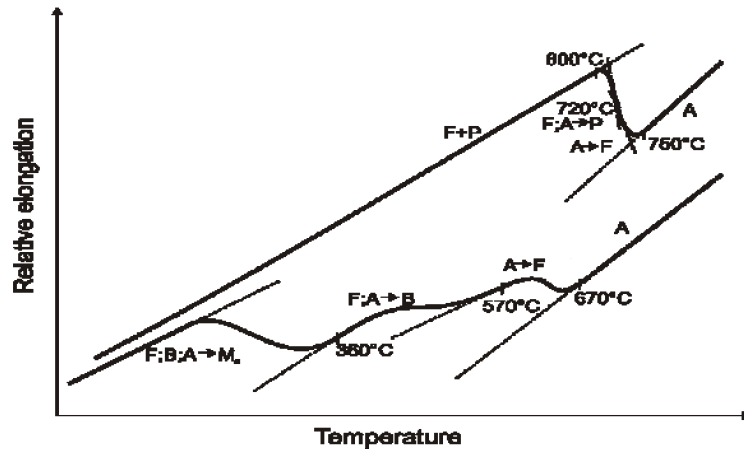

Figure 1: Schematic dilatation curves (A=austenite; F=ferrite; P=pearlite; B=bainite; M=martensite) [SEP1680].

2.2. The inverse problem

As described above, a dilatometer experiment provides two sets of time-dependent data: the length change, $\lambda(t)$, and the temperature at the middle point of the specimen, $\tau(t)$. Parameterized with respect to time, these data define the so-called dilatation curve (cf. **Figure 1**), which allows the determination of the transformation start and end. To obtain the resulting phase fractions in case of several product phases, an additional metallographic examination by a microscope is required.

For a cooling curve with at most two product phases, e.g., bainite and martensite, if it is assumed that their phase fractions are spatially homogeneous within the specimen, then $z(t) = (z_1(t), z_2(t))$ is only a function of time. From a mathematical point of view, it should be possible to obtain these two time-dependent functions from the measured data $(\lambda(t), \tau(t))$. Indeed it has been shown in [4] that the temporal evolution of the two phase fractions is uniquely determined by these two time-dependent measurements.

In other words, in case of at most two coexisting product phases, it should be possible to obtain all the information necessary to construct a CCT diagram directly from the dilatometer experiments, thereby avoiding additional metallographic investigations.

For the numerical identification of the phase fractions $z(t) = (z_1(t), z_2(t))$ from dilatation curves, the thermoelastic system (1a-d) is discretized with finite differences. The phase

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fractions are described as cubic splines. Enforcing additional assumptions $z'(0) = z'(t_E) = 0$, where t_E is the ending time, the remaining spline coefficients can be uniquely represented in terms of the z values in the temporal grid points, in the following called $\alpha_1, \cdots, \alpha_n$. Then the inverse problem is to find the values $\alpha_1, \cdots, \alpha_n$ such that the overall displacement $u(L,t)$ is close to the measurement $\lambda(t)$ and the temperature in the middle of the specimen, $T(L/2,t)$ is close to the measurement of $\tau(t)$ concurrently. More accurately, the following inverse problem has to be solved:

$$(IP) \quad \begin{cases} \min_{\alpha_1, \dots, \alpha_n} \left\{ \frac{\mu_1}{2} \int_0^{t_E} \left(u(L,t) - \lambda(t) \right)^2 dt + \frac{\mu_2}{2} \int_0^{t_E} \left(T(L/2,t) - \tau(t) \right)^2 dt \right\} \\ \\ \text{such that} \quad (u,T) \quad \text{solves} \quad (1a-d) \end{cases}$$

(IP) is a quadratic optimization problem with linear equality constraints, which can be solved, e.g., by the MATLAB Levenberg-Marquardt routine. To obtain useful results, the equilibration of both terms in the cost functional is indispensable.

2.3. Identification based on model data

In the first step, the validity of this approach was proven by using model data from the plain carbon steel SAE C 1080 (0.8%C, 0.76%Mn). Thus, further technical difficulties arising in connection with the evaluation of real dilatometer experiments were avoided. To generate the model data, the thermoelasticity system (1a-d) coupled with two rate laws for the phase fractions of pearlite and martensite was solved (cf. [4]).

In this study, only an interesting case of moderate cooling is presented, which yields two product phases: pearlite and martensite. Further details are referred again in [4].
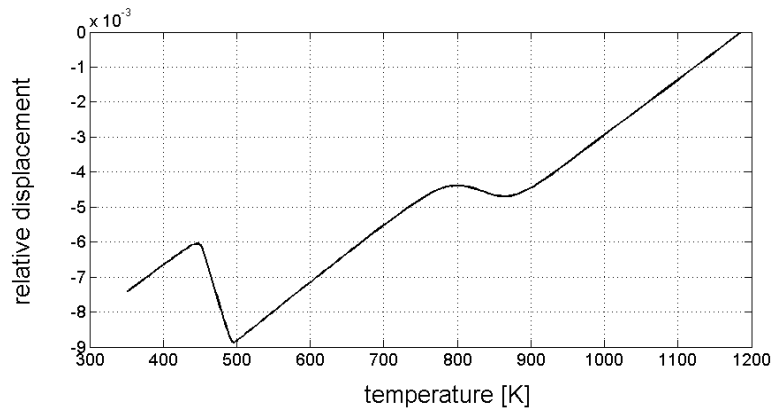

Figure 2: Model dilatometer curve for moderate cooling.

Figure 2 shows the model dilatation curve for the case of moderate cooling, exhibiting two phase transformations. **Figure 3** shows three iterations and the final result of the optimization process in this case. Starting from the initial values $z_1(0) = z_2(0) = 0$, the correct final phase fractions were already reached after three iterations.
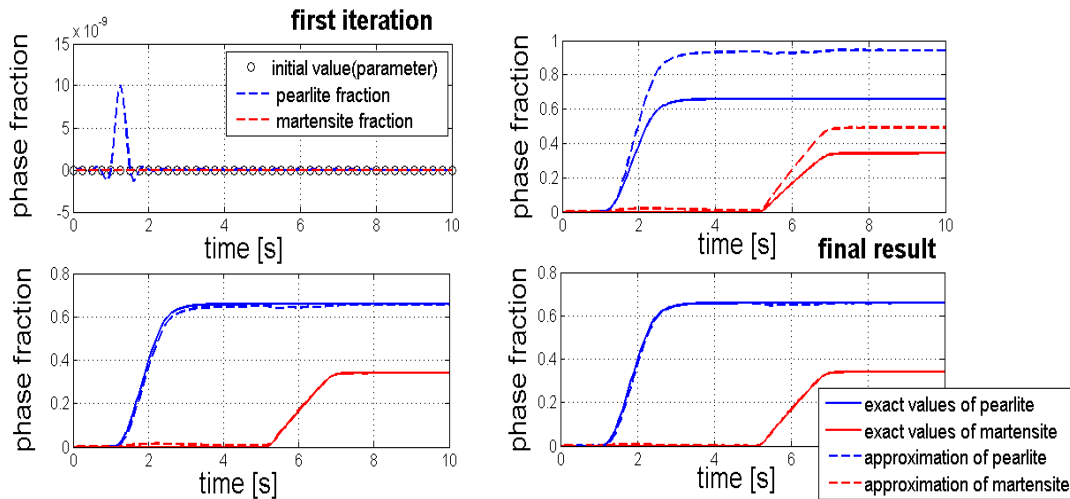
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Figure 3: Four iterations to compute phase fractions for moderate cooling.

3. Experimental Setup

The dilatometer tests were accomplished by a deformation dilatometer type DIL-805A/D fabricated by the company Baehr Thermoanalyse GmbH (**Figure 4**). It allows simulating many thermomechanical processes, as the machine has the ability to control heating and cooling rates up to 600 K/s, a maximum strain of 1.2 and a maximum strain rate of 13 s^{-1}. Hence, it is possible to investigate the transformation behaviour as a result of cooling rate and prior deformation.
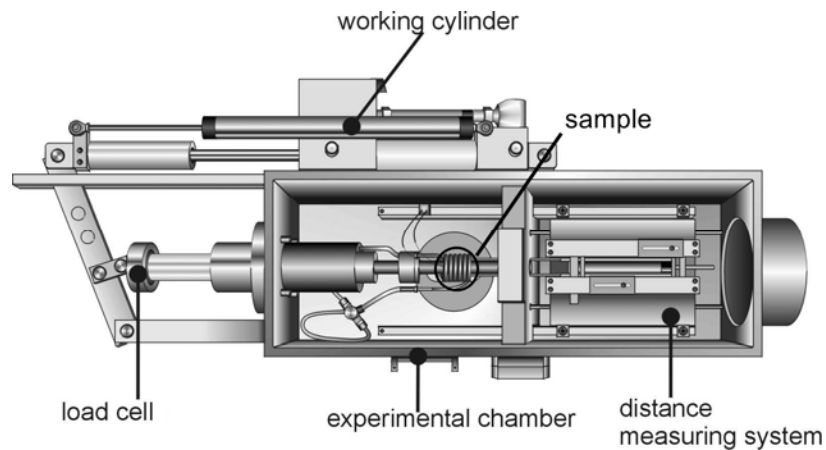

Figure 4: Schematic of the dilatometer.

From an elongation curve during continuous cooling as shown schematically in **Figure 1,** the beginning of the phase transformation is defined by the diversion of the elongation-time-curve from the straight line. The transformation is finished, when the curve turns into a straight line again. If there are successive phase transformations occurring in the steel, the inflection points are to be detected as the boundary between the transformation ranges.

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The dilatometer tests were performed according to SEP 1681 [10], while the results were converted into CCT-diagrams according to SEP 1680 [11].

In addition to the detection of the elongation curves, metallographic characterization was carried out in order to determine the amount of the different resulting phase fractions.

4. Results and Discussion

4.1. Experimental procedure

A medium carbon steel with chemical composition shown in **Table 1** was chosen in order to validate the numerical approach.

C	Si	Mn	P	S	Cr	Mo	Ni	Al
0.175	0.159	1.133	0.014	0.022	1.02	0.017	0.10	0.02

Table 1: Chemical composition of the steel 16MnCr5 in mass %.

Cylindrical dilatometer samples ($\varnothing$=5 mm, L=10 mm) were heated up to an austenitizing temperature of 1200 °C at a heating rate of 200 K/min, where the samples were austenitized for 10 minutes and then cooled down to room temperature with different cooling rates.

The phase transformation kinetics from the evaluation of the elongation curves, assisted by metallographic results, is presented in the CCT-diagram shown in **Figure 5**. The A_{c1} and A_{c3} temperatures of the steel 16MnCr5 were determined to be 740 °C and 835 °C respectively, while the martensite start temperature M_s was determined to be 400 °C. Additionally, the Vickers hardness test HV10 was performed for every cooling rate.
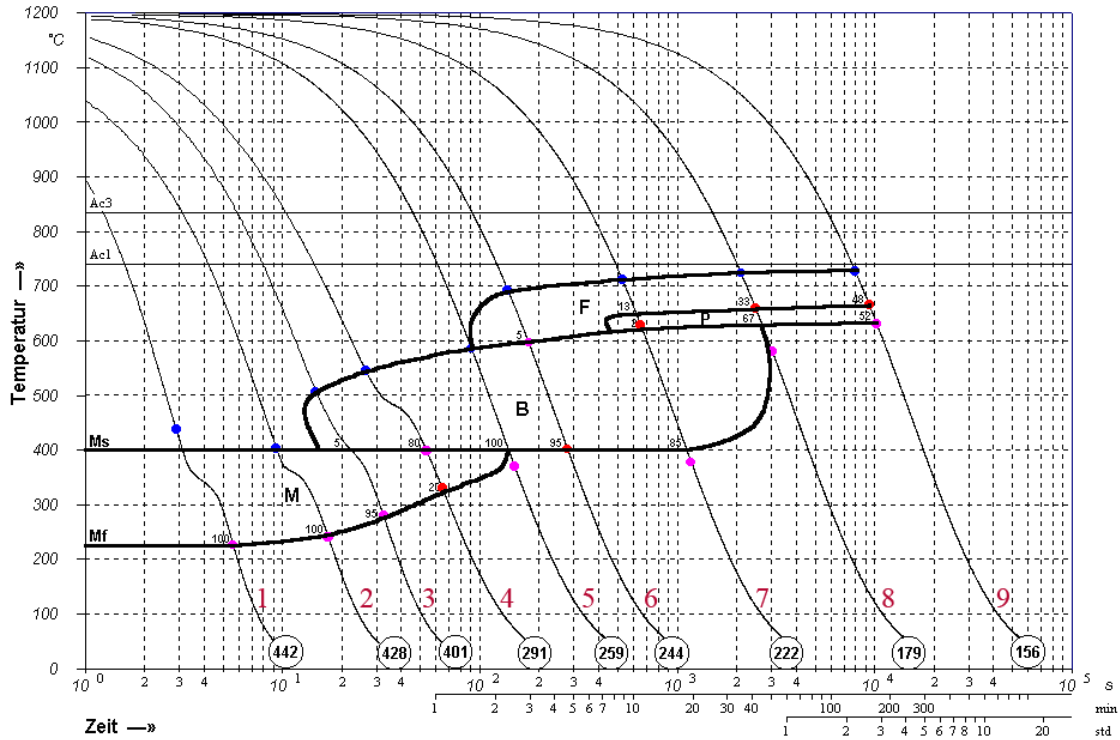
Figure 5: CCT-diagram of the steel 16MnCr5.

4.2. Input data for the numerical identification

Some published thermophysical data needed in equations (1a,b) according to the selected chemical composition i.e. thermal conductivity, density as well as heat capacity can be found in [12, 13]. Nonetheless, it is considerably complicated to acquire the others; the availability of the latent heats of the phase transformation comes from few references e.g., [14, 15]. More difficulties were found in case of thermal expansion coefficients and the reference temperatures in the mixing rule of the thermal strain.

The data for the Newton-type cooling model on the right-hand side of (1b) were also identified at the end. An appropriate measurable coolant temperature T^{ext} would be the temperature at the inner boundary of the tube surrounding the specimens in the dilatometer (cf. **Figure 4**). However, for preliminary technical reasons, T^{ext} was assumed to be constant.

In case of the heat transfer coefficient, or its dimensionless form, the Nusselt-number, many expressions for different geometrical configurations exist, e.g., [16]. However, none of them fit well with the dilatometer being considered. Typically, the Nusselt number is expressed as a product of further dimensionless characteristic numbers with empirical exponents. For the cooling curves c1 to c9 in **Figure 5,** different cooling strategies were used. For the highest cooling rates, i.e., curves c1 to c3, the coolant was blown onto the specimen (forced convection), while the cooling curves c4 to c9 were regarded as natural

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convection. Hence, the heat transfer coefficient had to be determined separately for these cases.

To decelerate the cooling in curves c5 to c9, the specimens were heated by electromagnetic induction. Although the Nusselt number was unaltered, this effect had to be taken into account by including an additional source term on the right hand side of the energy balance (1*b*). The electric power was measured for each cooling curve and a part of it was absorbed by the specimen. However, a precise model of this effect requires further studies which are beyond the scope of this paper. Therefore, these curves will be neglected in the sequel.

At first, the dilatometer data needed some modification; the dilatation curves corresponding to cooling curves c1 to c9 should have been consistent at the austenitic high temperature range. In other words, they all should have had similar slope. Nevertheless, it was not the case as the dilatation curves at high cooling rates differed from the ones with slow cooling rate. This observation can be interpreted as an inertia effect of heating. The temperature was measured at the surface of the specimen with a diameter of 5 mm. If the cooling rate is rather high, the temperature can differ from the inner temperature of the specimen. The temperature, T, becomes homogeneous in the specimen only under low cooling rates.

Taking into account of this fact, the data were converted in such a way that all cooling curves had the same displacement and slope in the high temperature range. As a reference, the cooling curve with the lowest cooling rate was used. This method should at least reduce the error due to the radial inhomogeneous temperature distribution inside the specimen.

4.3. Numerical results

In this procedure, the inverse problem algorithm was applied to the converted experimental data. Owing to the conversion, the thermal expansion coefficient and the reference temperature for austenite stay the same for all the cooling curves and can be drawn directly from the dilatation curves. As shown in **Figure 5**, the cooling curves c1 and c2 exhibit only one phase transformation i.e. from austenite to martensite. Hence, the thermal expansion coefficient and the reference temperature for martensite can be derived from the dilatation curves. The two values from both curves coincide with a relative error of merely less than one percent. Thus, all the data necessary to solve the inverse problem (IP) are available. **Figure 6** depicts the resulting evolution of martensite for c2.
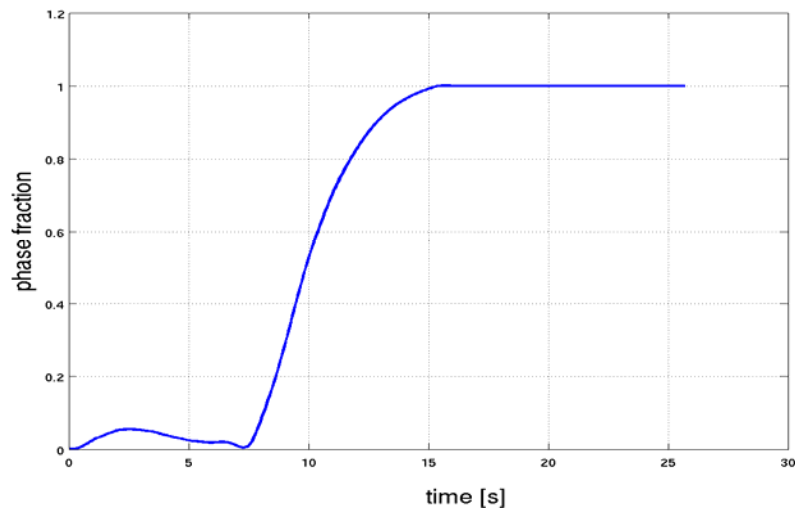
Figure 6: Time evolution of martensite for cooling curve 2.

A comparison with **Figure 5** shows that the martensite start and end temperatures have been captured quite precisely. The non-monotone behaviour at the beginning might be caused by the spatial one-dimensional model, which is particularly insufficient for the initial stage of cooling.

The next example is cooling curve c4, by which the metallographic investigations estimate 80% bainite and 20 %.martensite. To apply the algorithm to c4, the thermal expansion data for bainite must be searched for as they could not be drawn directly from the dilatation curve corresponding to c4. However, a close inspection of the dilatometer data revealed that cooling curve c5 exhibited only one phase transformation, which was assumed to be contributed by bainite. Thus, the thermal expansion coefficient and reference temperature for bainite could be drawn from curve c5. Consequently, all the data required for the solution of (IP) for cooling curve c4 are available.

Due to the fact that two product phases exist, the optimization procedure encountered additional difficulties. Therefore, some iterations of the convergence history in this case are depicted in **Figure 7**.
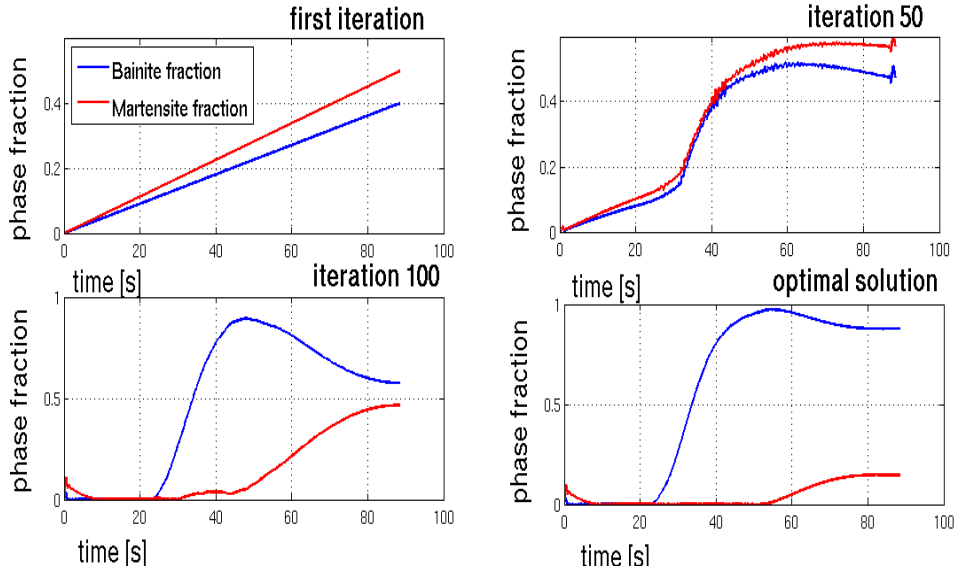
Figure 7: Some iterations to compute phase fractions for cooling curve 4.

The final phase fractions obtained by the optimization procedure are 85 % bainite and 15% martensite, which roughly correspond to the metallographic investigation. The evolution of martensite shows a satisfactory monotonic behaviour.

Beside a small non-monotonicity at the beginning of the bainite curve, there was also an overshooting before the approximately correct final phase fraction was reached.
It was found that the residual of the cost functional in (IP) was smaller in this solution than the one corresponding to a monotonic approximation of the bainite curve. This may indicate further errors in the data used for the numerical simulations, especially the latent heat, and is subjected to further studies.

The algorithm applied to cooling curve c3 yields 100% martensite with no bainite, giving the same error interval as the result for cooling curve c4. **Table 2** lists the obtained numerical values for the thermal expansion coefficients and reference temperatures for austenite, bainite and martensite. The reference temperatures depend on the chosen reference which is the length at 1000^oC in this case. Only the differences between the reference temperatures are material dependent.

phase	δ_i	T_i^{ref}
austenite	$2.5156 \cdot 10^{-5}$	$1000.3 \ ^oC$
bainite	$1.5457 \cdot 10^{-5}$	$996.0 \ ^oC$
martensite	$1.3372 \cdot 10^{-5}$	$1170.0 \ ^oC$

Table 2: Thermal strain data for austenite, bainite and martensite.

5.   Summary and Conclusion

The goal of this paper is to review and extend the evaluation methodology of dilatometer experiments. A mathematical analysis of the underlying thermomechanical model shows that in the case of at most two product phases, the complete transformation kinetics of these phases should be uniquely determined by the two dilatometer measurements of length-change and temperature evolution in a single point on the specimen surface.

Stimulated by this conclusion, an inverse problem was formulated to identify the evolution of phase fractions via the solution of an optimization problem. When applied to the model data, the method exhibits a fast convergence to the exact solution.
A dilatometer experiment for the steel 16MnCr5 was conducted. Aided by additional metallographic investigations, a complete CCT diagram was derived in a standard way. Then the numerical identification method was applied to this steel. In case of a single phase transformation, the kinetics could be completely determined while in case of two product phases, the final phase fractions could be estimated with an absolute error of 5% compared with the metallographic results.

Further investigation is still needed for the final judgement on the advantages of the new approach described in this paper. The results so far have clearly demonstrated that the dilatometric data contain plenty information rather than simply the start and end temperatures of the related phase transformations. Furthermore, the improvement of the mathematical model of the dilatometer experiments has been planned.

A refinement of the presented method including a 2-stage optimization algorithm for the consecutive coefficients identification for the thermal strain model and the identification of phase transformation kinetics probably allows excluding the metallographic analyses in the dilatometer experiment evaluation completely.

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