

## Mathematical modelling of NbC particle nucleation and growth in an HSLA steel under elastic deformation

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**Abstract.** This study attempts to incorporate the effect of elastic deformation in a previously proposed model for the nucleation and growth of precipitates. We adapt the KWN-model by Robson to incorporate the effect of strain energy arising from elastic deformation on the homogeneous nucleation and growth of NbC particles in a High Strength Low Alloy (HSLA) steel at constant temperature. Simulations of the nucleation and growth of NbC particles in an HSLA steel on the cylindrical region show that the incorporation of elastic strain energies has a noticeable impact on the process. The derived quantities of homogeneous nucleation and growth, such as the particle number density and the mean particle radius, show a clear spatial correlation with the calculated strain energy.

### Introduction

Metalworking of alloys is a complex operation that has several influential factors on the usability of the alloy. The influence of most of these factors has been studied and achieved by a process of trial and error. An analytical approach to investigate these factors could verify the obtained experimental results and extend the knowledge about the behaviour of alloys during metalworking.

During the last two decades various models for the nucleation and growth of precipitates in alloys have been proposed and evaluated. These models can be divided in four general categories. The first category concerns models which predict the nucleation and growth of precipitates on the evolution of a particle size distribution function. A classical example of this model is the KWN-model by Kampmann and Wagner [1], which has been extended and evaluated by, amongst others, Robson [2]. The second category consists of models that predict the nucleation and growth of precipitates using a mean variable approach, which model the time evolution of variables such as the mean particle radius and the particle number density. An example of such a model has been proposed by Deschamps and Brechet [3]. The next category contains models that predict the growth and dissolutions of precipitates present in a system by solving Stefan-like problems for different precipitate sizes. One of the latest models in this area is developed by Vermolen et al. [4]. Finally a category containing models which describe precipitation dynamics using statistical methodologies can be identified, such as the model by Soisson et al. [5].

This article will present an adapted KWN-model from the first category for homogeneous nucleation and growth and particles by Robson's formalism [2] to present a model for homogeneous nucleation which incorporates the effect of elastic deformations. This goal will be achieved by reducing the number of empirical parameters and introducing (neglected) energy terms. The elastic deformations will be modelled using the standard linear elastic models in cylindrical regions, see for example [6]. Next the model will be demonstrated by an application to an undeformed hypothetical HSLA steel and

subsequently the results of the influence of a tensile test on a specimen will be given and discussed. The numerical methods that are applied to the model to be able to perform the simulations will be discussed in a future paper.

### The model

The nucleation and growth of precipitates are modelled in the current paper by the KWN model from Robson [2]. We present an extensive discussion of this model. The main features of the KWN model are:

- All particles are spherical and classified with their spherical radius in m.
- The time behaviour in seconds (s) of the model is described by the partial differential equation:

$$\frac{\partial N}{\partial t} = -\frac{\partial[Nv]}{\partial r} + S, \quad (1)$$

in which  $N \equiv N(r, t)$  in  $\text{m}^{-3}$  represents the number of particles with radius  $r$  at time  $t$ ,  $v \equiv v(r, t)$  in  $\text{ms}^{-1}$  the growth rate of particles with radius  $r$  at time  $t$  and  $S \equiv S(r, t)$  in  $\text{m}^{-3}\text{s}^{-1}$  a source function predicting the number of new particles with radius  $r$  at time  $t$ .

- The value of the source function  $S$  is calculated from classical nucleation theory (CNT) and is given by

$$S(r, t) = \begin{cases} I(t) & \text{if } r \in [r^*(t), 1.05r^*(t)], \\ 0 & \text{otherwise.} \end{cases} \quad (2)$$

Here  $I \equiv I(t)$  is the nucleation rate of the particles following from CNT and  $r^* \equiv r^*(t)$  the critical radius following from CNT. The value 1.05 is adopted from Myhr and Grong [7].

In this paper, we couple the KWN-model with a standard model for linear elastic deformations, which will not be discussed here, but see for example [6].

**Nucleation of precipitates** Robson [2] assumes that the time-dependent homogeneous nucleation rate  $I$ , as used in (2), is given by

$$I = N_v Z \beta^* \exp\left[-\frac{\Delta G^*}{kT}\right] \exp\left[-\frac{\tau}{t}\right], \quad (3)$$

where  $k$ ,  $T$  and  $t$  represent the Boltzmann constant, the absolute temperature and time. Furthermore,  $N_v$  is the number of potential homogeneous nucleation sites per unit volume,  $Z$  the Zeldovich factor,  $\beta^*$  the atomic attachment to a growing particle and  $\tau$  the incubation time for homogeneous nucleation. For the formulae of the latter three variables, we refer to [2]. The term  $\Delta G^*$  is the free energy barrier for homogeneous nucleation which must be overcome before precipitation occurs.

In the classical nucleation theory, the free energy for homogeneous nucleation  $\Delta G$  is assumed [8] to be of the form

$$\Delta G = \frac{4}{3}\pi r^3 \Delta G_v + 4\pi r^2 \gamma, \quad (4)$$

for coherent spherical particles, of which the derivative with respect to  $r$  is equated to zero and solved for  $r$  to find the critical radius  $r^*$  and the corresponding free energy barrier for homogeneous nucleation  $\Delta G^*$ . In this formula  $\Delta G_v$  is the chemical volume free energy density and  $\gamma$  the surface energy of the precipitate, which will be discussed below. We adapt equation (4) by assuming that the free energy for homogeneous nucleation of a precipitate with radius  $r$  is reduced due to the releasing of elastic strain

energy in the matrix. Assuming the external elastic strain energy density  $\Delta G_s^{el}$  to be known, the free energy  $\Delta G$  can be redefined by:

$$\Delta G = \frac{4}{3}\pi r^3(\Delta G_v - \Delta G_s^{el}) + 4\pi r^2\gamma. \quad (5)$$

In the above formula it is assumed that  $\Delta G_v$  has negative sign, as originally derived in [9], and  $\Delta G_s^{el}$  has positive sign. The latter can be shown by assuming that a material is deformed linear elastically in some manner, resulting in a strain tensor and a strain tensor, which will have similar signs due to linearity. As a result the elastic strain energy density  $\Delta G_s^{el}$  will have in all cases a positive sign, thereby increasing the free energy for homogeneous nucleation. Differentiation Equation 5 with respect to  $r$  and equating to zero gives the new critical radius and the corresponding homogeneous nucleation energy barrier

$$r^* = \frac{-2\gamma}{\Delta G_v - \Delta G_s^{el}} \quad \text{and} \quad \Delta G^* = \frac{4}{3}\pi\gamma(r^*)^2. \quad (6)$$

Following the proof and assumptions given by Aaronson [9] for the volume free energy density for a binary system, the volume free energy density for any alloy can be expressed by

$$\Delta G_v = -\frac{RT}{V_m^p} \sum_{i=1}^n x_i^p \ln \left( \frac{x_i^m}{x_i^e} \right), \quad (7)$$

where  $V_m^p$  is the molar volume of the precipitate,  $x_i^p$  the molar fraction of element  $i$  in the precipitate,  $x_i^m$  the molar fraction of element  $i$  in the matrix,  $x_i^e$  the equilibrium molar fraction of element  $i$  in the matrix and  $R$  the gas constant and  $n$  total number of elements, including the solvent. The equilibrium molar fractions and related weight percentages are determined using a known solubility product and an equilibrium mass balance, also known as the lever rule.

On the value of  $N_v$ , the number of potential homogeneous nucleation sites per unit volume, see equation (3), various theories are present. For a discussion on the value of  $N_v$  we refer to Robson [2], who suggests using the number of solute atoms per unit volume as value for  $N_v$  which can be adjusted by assuming the solute molar fraction as empirical parameter. Instead of using the molar fraction as an empirical parameter, we use the following formula for the number of potential homogeneous sites  $N_v$

$$N_v = \min_{\{j: x_j^p \neq 0\}} \frac{x_j^m}{V_m^m} \frac{V_m^p}{\zeta_j^p V^*}, \quad (8)$$

in which  $V_m^m$  the instantaneous molar volume is of the matrix,  $\zeta_j^p$  the number of atoms of element  $j$  in a single particle molecule and  $V^*$  the volume of a single particle with radius equal to the critical radius  $r^*$ .

## Growth

In the previous section the nucleation rate  $I$  has been discussed, which is incorporated in the source function  $S$  of equation (1). The other factor influencing the time-evolution of the particle distribution is the growth rate  $v$ . Following Myhr and Grong [7], we choose a growth driving element  $P$  from the elements present in the particle and set

$$v = \frac{D_P}{r} \frac{C_P^m - C_P^i}{C_P^p - C_P^i}, \quad (9)$$

in which  $C_P^m$  is the weight percent of solute in the matrix,  $C_P^i$  the weight percent of solute in the matrix at the particle/matrix interface,  $C_P^p$  the weight percent of solute in the particle and  $D_P$  the diffusion coefficient of element  $P$  in the matrix. The value of the concentration  $C_P^i$  is modelled by an application of the Gibbs-Thomson equation

$$C_P^i = C_P^e \exp\left(\frac{2\gamma V_m^p}{RT} \frac{1}{r}\right). \quad (10)$$

We assume at this moment that the value of the particle/matrix interface energy  $\gamma$  is the same for both the growth of particles as the nucleation of particles, which is in line with [2].

## Application of the model

The above derived model is applied using standard available numerical methods to a hypothetical HSLA steel containing 0.017Nb, 0.06C and 0.25Mn (in wt%). It has been held at 1250 degrees Celsius for a long period of time with a fully austenitic structure and then fast cooled to 950 degrees Celsius, at which the simulations start. It is assumed that only NbC precipitates can occur, no precipitates are initially present and Nb is the element driving growth of precipitates. Furthermore it is assumed that the surface energy  $\gamma$  has a value of  $0.15 \text{ J/m}^2$ . For the solubility product that we needed to obtain the equilibrium concentrations and the diffusivity of Nb in austenite we follow Dutta et al. [10] giving

$$\log_{10} C_{Nb}^e C_C^e = 2.06 - \frac{6700}{T} \quad \text{and} \quad D_P = 1.4 \times 10^{-4} \exp\left(-\frac{270 \times 10^3}{RT}\right). \quad (11)$$

Figure 1 gives some results of the model when applied to the HSLA steel prior to deformation. These results show that initially small precipitates nucleate with an increasing nucleation rate, causing a rapid increase in the particle number density. The nucleated precipitates grow immediately after nucleation, causing a simultaneous increase in mean particle radius. After about  $8 \times 10^4$  seconds the nucleation rate drops to almost zero, causing a stabilisation of the particle number density. As the system has not attained an equilibrium, which can be seen from the values of the Nb matrix concentration, the present precipitates will start to coarsen until the equilibrium is almost achieved. At this moment the Nb matrix concentration is close to the equilibrium value of 0.0065 wt%.

Figure 2 shows a typical elastic strain energy density distribution in a ASTM standard specimen following page 6 from [11] during a tensile test. The value of the elastic strain energy is determined using an implementation of a model for linear elastic deformations ([6]). This energy distribution was used as input parameter for the elastic energy in equation (5). Simulating the nucleation and growth in the mentioned HSLA under deformation, the result in Figures 3, 4 and 5 are obtained. These results depict the results under elastic deformation as percent differences with the results in Figure 1 at various moments during simulation.

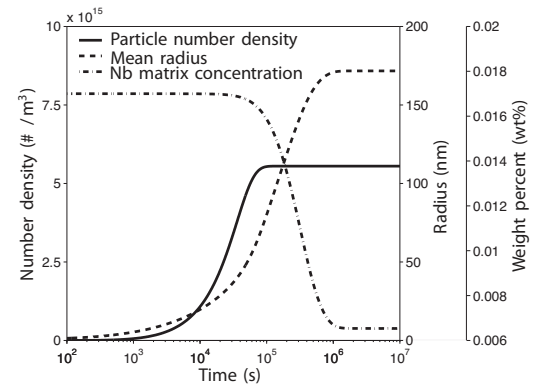


Fig. 1: Particle number density, mean radius and Nb-concentration in the matrix as function of time at zero stress.

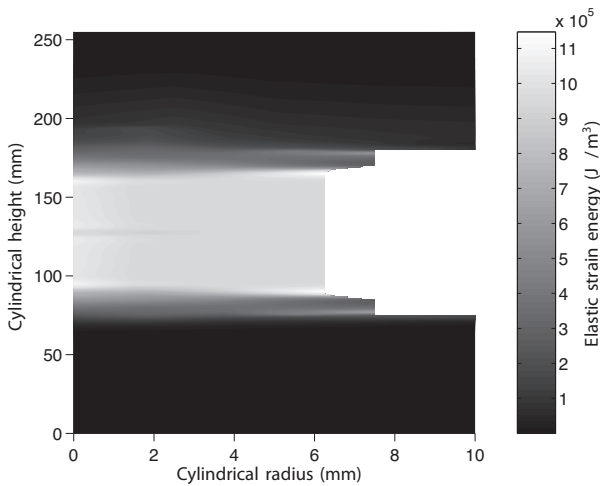


Fig. 2: Simulated elastic strain energy distribution on a cylindrical specimen during simulations.

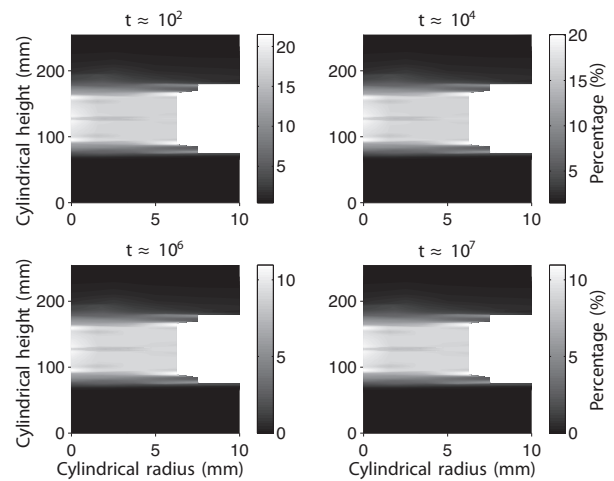


Fig. 3: Relative differences of the local particle number density at zero and near 550 MPa stress.

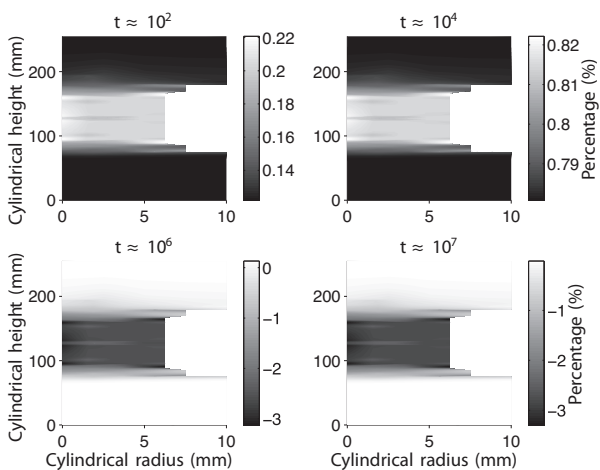


Fig. 4: Relative differences of the local mean particle radius at zero and near 550 MPa stress.

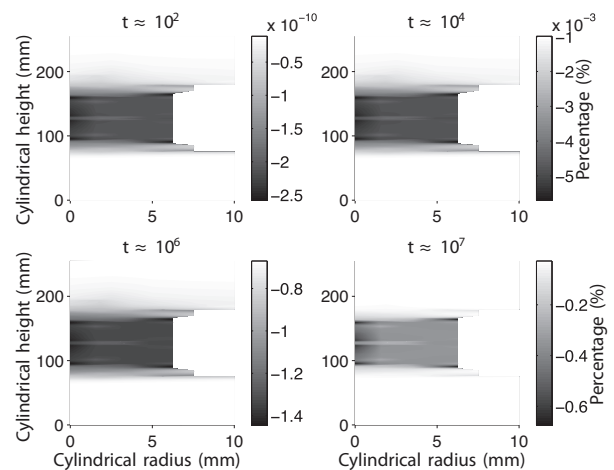


Fig. 5: Relative differences of the local Nb concentration in the matrix at zero and near 550 MPa stress.

The results indicate that the introduction of elastic strain energy as proposed in the model causes a significant increase in the particle number density throughout the simulation with a clear spatial correlation with Figure 2. From Figure 4 we can derive that during the initial stages of the model larger particles nucleate with respect to the results without deformation, thereby increasing the mean particle radius. As soon as the particle number density stabilizes, the precipitates will start to grow. Due to the higher particle number density with respect to the undeformed system, less niobium is present in the matrix to facilitate the coarsening. This indicates that the precipitates can not become as large as in the undeformed system, causing a relative lower increase in the mean radius, which in turn leads to a relative lower mean radius. Due to the correlation between the particle number density (Fig. 3) and the strain energy (Fig. 2) the mean radius also shows a clear correlation with Figure 2. The results for both the mean particle radius and the particle number density indicate that the concentration of niobium in the matrix should be lower at the initial stages of the model, but this should eventually cancel out as equilibrium is attained, which can be seen back in Figure 5.

## Conclusion

We simulated the effect of external elastic deformations on the nucleation and growth of homogeneous NbC precipitates in an HSLA steel. The simulations show a successful introduction of the external elastic strain energy in the KWN-model and that this introduction can lead to more and larger NbC precipitates at the start of the process. At the end of the process the system will contain more precipitates with a smaller mean radius.

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## **Mathematical Modelling of NbC Particle Nucleation and Growth in an HSLA Steel under Elastic Deformation**

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