# CELLULAR AUTOMATA MODEL OF CARBONITRIDE PRECIPITATION PROCESS TO SIMULATE IMAGE OF MICROSTRUCTURE IN MICROALLOYED STEELS

MARYNOWSKI Przemysław[1] ADRIAN Henryk[1] GŁOWACKI Mirosław[1,2] WOŹNY Krzysztof[1]

[1]*AGH University of Science and Technology, Cracow, Poland, EU*

[2]*The Jan Kochanowski University (JKU), Kielce, Poland, EU*

**Abstract**

Proposition of a Cellular Automata (CA) model of carbonitride precipitation to simulate image of microstructure in microalloyed niobium steels is presented in the paper. CA model proved to be very efficient in modeling various phenomena in material science. Numerical modeling played important role in development of new processing technologies taking advantage of precipitation. By modeling we mean a mathematical description of the relation between the main process variables and the resulting material properties, based on sound physical principles. In microalloyed steels the microalloying elements: Ti, Nb, V are added in order to control their microstructure and mechanical properties. High chemical affinity of these elements for interstitials (C, N) result in precipitation of binary compound, nitrides and carbides and products of their mutual solubility - carbonitrides. The model accounts for an increase of dislocation density due to plastic deformation and predicts kinetics of precipitation as well as shape of precipitates.

**Keywords:** Cellular automata, carbonitrides, microalloyed niobium steels, microstructure

## 1. INTRODUCTION

Understanding and modeling microstructure evolution is a major concern in the scientific and engineering fields. Due to the difficulty of directly incorporating topological features into mathematical models, together with the difficulty of providing a space-time description, there has been increasing interest in using computer simulation to study and predict the microstructure evolution in a range of technologically important materials [1]. Most of the thermodynamics models were developed in the second half of the 20th century. For example the model of carbonitrides precipitation in microalloyed steels of Dutta & Sellars [2], Dutta et al. [3], Dutta et al. [4]. The models allow calculate kinetics of precipitation and changes of the size of precipitates as a function of processing parameters. In the present paper the simplified model accounting for one alloying element, niobium [5], is used in CA method to precipitation process to get image of the microstructure. The main advantage of the model is simulation of the microstructure image for given chemical composition of steel and parameters of technological process. The CA method offers a reasonable balance between its computational simplicity and ability to provide quantitative results.

## 2. PHYSICAL BASE OF PRECIPITATION IN MICROALLOYED NIOBIUM STEELS

Carbonitride precipitations has played crucial role in controlling of mechanical properties of microalloyed steels. The one of microalloying element Nb is added into low alloyed steels to control their microstructure and mechanical properties. High chemical affinity of those elements for interstitials (C, N) results in precipitation of binary compound, nitride and carbide and products of their mutual solubility - carbonitride [6]. The composition of carbonitrides depends on the composition of steel as well as on the temperature. For calculation of the chemical composition of austenite, the composition and volume fraction of carbonitrides the thermodynamic models were developed [5, 7, 8, 9], based on the regular solution model for stoichiometric phases developed by [10]. The effect of addition of Ti, Nb and V in low alloyed steels on the thermodynamic equilibrium state is described by Adrian model [5, 11].

The thermodynamic model allows to calculate the chemical composition of austenite, the composition and volume fraction of carbonitrides at given temperature under thermodynamic equilibrium conditions based on the knowledge of the chemical composition of the steel. The thermodynamic model does not allow for the calculation of the size of precipitations. Information about the size of precipitations produced under non-equilibrium thermodynamic conditions enables the models of kinetics of carbonitride precipitation [12-17].

The non-equilibrium thermodynamic state is usually obtained by decreasing of the temperature or by an increase of the pressure. The way of disintegration of the primary phase depends on a number of parameters and factors and is difficult to predict. Some of this information was used in the present work to develop the transition rules for the CA model to predict the image of microstructure in microalloyed niobium steels.

## 3. CA MODEL

CA is mathematical tools that can be used to model physical systems. The main principles of the applications of the CA method in materials science were discussed by Raabe [18]. Modelling microstructure evolution is the most frequent application of the CA [19].

Idea of the CA technique is to divide a specific part of the material into one-, two-, or three-dimensional lattices of finite cells. Each cell in this CA space is called a cellular automaton, while the lattice of the cells is known as cellular automata space. Each cell is surrounded by neighbours, which affect one another. Neighbourhoods can be specified in one-, two-, and three-dimensional spaces. The most popular examples are the von Neumann and the Moore neighbourhoods [18], where in the 2D case each cell is surrounded by either four or eight neighbouring cells, respectively.

Each cell in the CA space is characterised by its state and by values of internal variables. The cells interactions within the CA space are based on the knowledge defined while studying a particular phenomenon. In every time step, the state of each cell in the lattice is determined by the previous states of its neighbours and the cell itself on a basis of a set of precisely defined transition rules:

$$Y_{i,j}^{t+1} = \begin{Bmatrix} if\ (\Lambda) \Longrightarrow newstate \\ else\ \Longrightarrow\ Y_{i,j}^{t} \end{Bmatrix} \tag{1}$$

where: $Y_{i,j}^{t+1}, Y_{i,j}^{t}$ - state of the cell *i,j* in the current and previous time step, respectively, $\Lambda$ - logical function, which describes the condition when the state of the cell changes. Function $\Lambda$ depends on:

$$\Lambda = \Lambda\left(Y_{i,j}^{t}, Y_{k,l}^{t}, \boldsymbol{p}, \boldsymbol{q}\right) \tag{2}$$

where: $Y_{k,l}^{t}$ - state of the cell *k,l,* which is a neighbour to the cell *i,j*, in the previous time step, ***p*** - vector containing external variables, e.g. temperature, ***q*** - vector containing internal variables, e.g. carbon concentration.

Since the transition rules control the cells behaviour during calculations (i.e., during the deformation and precipitation processes), the proper definition of these rules in designing a CA model critically affects the accuracy of this approach. The transition rules of the developed CA model of precipitation are based on the knowledge of experts, scientists, experimental observations, and available literature knowledge.

## 4. CA PRECIPITATION MODEL

Publications dealing with modelling of precipitation using the Cellular Automata are scarce. Objective of the present work was to apply CA technique to simulate strain induced transport of carbon and niobium in steel and further formation and growth of carbonitride precipitates and predict the image of microstructure in microalloyed niobium steels.

Conventional equations describing precipitation of carbonitrides, which were used in building the transition rules, are given below. Time between the deformation and beginning of precipitation was calculated as [2]:

$$t_{0.005P} = \frac{3\ x\ 10^{-6}}{[Nb]\varepsilon\sqrt{Z}} exp\left(\frac{270000}{RT}\right) exp\left\{\frac{2.5\ x\ 10^{-10}}{T^3[\ln(k_s)]^2}\right\} \quad (3)$$

where: $\varepsilon$ - strain, $Z$ - Zener-Hollomon parameter, $R$ - gas constant (J / (mol·K)), $T$ - absolute temperature (K), $k_s$ - supersaturation ratio, being the ratio of actual amount [Nb] [C + (12/14)N] to the equilibrium amount, defined as:

$$k_s = \frac{[Nb]\left([C]+\frac{12}{14}[N]\right)}{10^{\frac{6770}{T}-2.26}} \quad (4)$$

Critical radius for nucleation, $r_{cr}$, which is assigned to a new nucleus in the CA model, is determined by the driving force and the equation is:

$$r_{cr} = -\frac{2\gamma}{\Delta G_v} \quad (5)$$

where: $\gamma$ - energy of the interface equal to 0.5 J / m$^2$, $\Delta G_v$ - the difference in free energy per unit volume (J / m$^3$), which is calculated from the formula:

$$\Delta G_v = -\frac{RT}{V_m} ln(k_s) \quad (6)$$

where: $V_m$ - molar volume; for Nb(C,N), $V_m$ = 1.28 x 10$^{-5}$ m$^3$ / mol.

At elevated temperatures coagulation of the particles of precipitates occurs, what is described by the following equation:

$$r^3 - r_0^3 = \frac{8}{9}\frac{D_{Nb}[Nb]\gamma V_m^2}{RT} t \quad (7)$$

where: $D_{Nb}$ - diffusion coefficient for niobium (m$^2$ / s), $t$ - time (s).

Deformation and resulting stresses has strong influence on precipitation. Dislocation density is used in the model as a measure of deformation. Increment of the dislocation density in a time step is calculated from the differential equation written in a differential form:

$$\Delta\rho = \left[\frac{\dot{\varepsilon}M}{bl} - B_0\dot{\varepsilon}^n exp\left(\frac{Q_s}{RT}\right)\rho\right]\Delta t \quad (8)$$

where: $t$ - time, $\dot{\varepsilon}$ - strain rate (1 / s), $M$ - Taylor constant, $b$ - length of the Burgers vector (2.59 x 10$^{-10}$ m), $l$ - average free path for dislocations (m), $n$ - coefficient, $B_0$ - coefficient of recovery, $Q_s$ - activation energy for self-diffusion (J / mol).

Presented equations are used in the model to calculate the conditions, which are used in the transition rules.

### 4.1. CA model - states of cells and variables

Two dimensional CA space was created. Two alternative neighbours were tested: Moore - as neighbors, assume all cells, the cells lying on the sides and in the corners, Von Neumann - as neighbors, take only the cells lying on the sides of the cell. Since dimensions of precipitates are few orders magnitude smaller than the grain size, the modelling process was carried out in a domain, which represented very small part of the material. Three possible states of the cell were introduced: *austenite* ($\gamma$), *precipitate* ($P$) and boundary ($\gamma$ - $P$). Beyond this, each cell was characterised by the internal variables: nucleation rate ($N$), dislocation density ($\rho$).

The following external variables were assumed: concentration of carbon, nitride and niobium in steel ([C], [N], [Nb]), current radius of the precipitate (*r*).

### 4.2. CA model - transition rules and flow of calculation

The transition rules transfer the mathematical model and the knowledge regarding precipitation into the CA space. The cell, which belongs to the austenite grain, will become a nucleus of a precipitate, with certain probability if:

- it has a dislocation density exceeding critical value $\rho_{cr}$, which is a function of the temperature (decrease of the temperature results in a decrease of a critical dislocation density).

The cell, which belongs to the austenite grain, will become a precipitate if

- it has at least one neighbour, which is a precipitate and the displacement of the $\gamma$ - *P* interface is larger than the distance between the cells and the content of MA in this cell is above equilibrium level.

The cell, which is a precipitate, will coagulate if

- it has at least one neighbour, which is also a precipitate and the increase of the radius *r* is larger than the distance between the cells.

In each time step calculations begin with determination of the increment of the dislocation density. This increment is distributed randomly between all the cell, except the cell which are *Precipitate*. Dislocations are allowed to migrate randomly but they cannot cross austenite grain boundaries. In consequence, random distribution of dislocation density is obtained with higher density close to the grain boundary and lower density inside the grains. Niobium is removed from the neighbour cells to the precipitate. The transition rules are checked at each time step. When deformation is finished, the dislocation density remains constant, what means that the model at this stage does not account for the recovery. This relation has to be added in each cell separately, because recovery depends on the current level of the dislocation density. Calculations are stopped when the content of niobium in steel is too low to form new precipitate cells.

In C# was written and implemented in the Visual Studio 2010 the MPCA program (Marynowski Przemysław Cellular Automata). Graphical interface was added. The following input data are introduced through the interface: content of niobium, carbon and nitrogen in steel, temperature, strain rate and size of the Cellular Automata space. The following parameters are calculated by the model: distribution of the dislocation density, distribution function for the size of precipitates and an average size of precipitates and composition of austenite. The model contains several parameters, which are not known a'priori. These parameters are critical dislocation density, niobium content necessary to create a precipitate.

## 5. RESULTS

Steel containing 0.2 % C, 0.02 % Nb and 0.015 % N subjected to heat treatment austenitization at 1200 °C with following holding at T for $\tau$ time with different strain was considered. Since precipitates are few orders of magnitude smaller than the austenite grain size, generation of the initial microstructure was limited to initiation of the input parameters. The results of image calculations of microstructure with carbonitride precipitations presented in **Figure 1** and **Figure 2** were obtained for the 300 x 300 cells, while the dimension of a single cell was 1 nm.

Increase of strains leads to rapid decrease of the precipitation time and to larger number of precipitates. It is due to the fact that occurrence of larger number of precipitates leads to a decrease of the niobium content in the austenite and, in consequence, the growth of precipitates is retarded.

Precipitation is thermally activated process and strong influence of the temperature is expected. Solubility of niobium in an iron decreases rapidly with a decrease of the temperature. Decrease of the temperature leads to a decrease of the average radius of precipitates and to an increase of the number of precipitates.

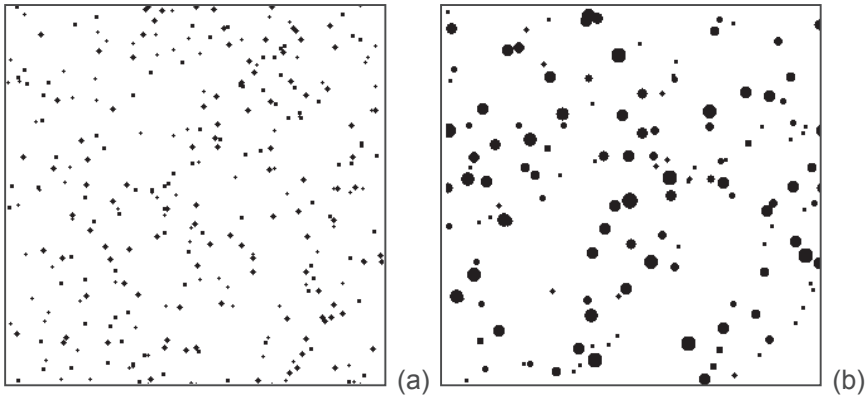


**Figure 1** Comparison of simulated microstructures with precipitations of carbonitrides in microalloyed niobium steel after heat treatment: austenitization at 1200 °C with following isothermal holding at 880 °C (a) and 980 °C (b) for 700 sec and strain 0.01.

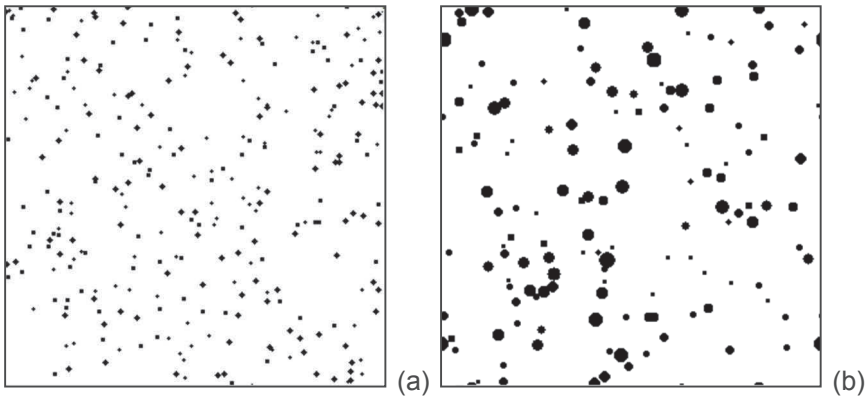


**Figure 2** Comparison of simulated microstructures with precipitations of carbonitrides in microalloyed niobium steel after heat treatment: austenitization at 1200 °C with following isothermal holding at 880 °C (a) and 980 °C (b) for 450 sec and strain 0.2.

## 6. CONCLUSION

Mechanical properties of microalloyed steels are determined by their chemical composition and technological process parameters, which have influence to the size of carbonitrides precipitations. Precipitations inhabit grain growth. Refining of grains allows to obtain high mechanical properties of steel after cooling after plastic working without additional heat treatment. Based on the chemical composition and size of the precipitations, the effect of microalloying element on mechanical properties of steels can be determined. A proposition of the CA model for the analysis of the precipitation of carbonitrides in microalloyed niobium steels was presented in the paper. The developed MPCA program allows to calculate the microstructure image from given process parameters (temperature and time) and chemical composition of steel.

The model needs further development, which should include:

- Identification of such parameters as critical dislocation density, the size distribution of precipitation and its effect on mechanical properties of steel
- Validation of the model by a comparison of predictions with the experimental data.

## ACKNOWLEDGEMENTS

***Work within the AGH project 11.11.110.300***

## REFERENCES

[1] GŁOWACKI, M., DĘBIŃSKI, T., OLEJARCZYK-WOŻEŃSKA, I.Fast Parallel Computation Algorithm in Application to Simulation of Semi-Solid Steel Rolling and Inverse Analysis of its Properties In *11th International Conference of Numerical Analysis and Applied Mathematics* 2013, Cracow: PAN, vol. 1558, pp. 2179-2182.

[2] DUTTA, B., SELLARS, C. M. Effect of composition and process variables on Nb(C,N) precipitation in niobium microalloyed austenite. *Materials Science and Technology*, 1987, vol. 3, pp. 197-206.

[3] DUTTA, B., VALDES, E., SELLARS, C. M. Mechanism and kinetics of strain induced precipitation of Nb (C, N) in austenite. *Acta Metallurgica et Materialia*, 1991, vol. 40, pp. 653-662.

[4] DUTTA, B., PALMIERE, E. J., SELLARS, C. M. Modelling the kinetics of strain induced precipitation in Nb microalloyed steels. *Acta Materialia*, 2001, vol. 49, pp. 785-794.

[5] ADRIAN, H. Thermodynamic model for precipitation of carbonitrides in high strength low alloyed steels up to three microalloying elements with or without additions of aluminium. *Materials Science and Technology*, 1992, vol. 8, pp. 406-420.

[6] GOLDSCHMIDT, H. J. *Interstitial Alloys*, London: Butterworths, 1967.

[7] LIU, W., JONAS, J. J. Calculation of the Ti(CyN1-y) - Ti4C2S2 - MnS austenite equilibrium in Ti-bearing steels. *Metallurgical Transactions A*, 1989, vol. 20A, pp. 1361-74.

[8] ROBERTS, W., SANDBERG, A. *The composition of V(C, N) as precipitated in HSLA steels microalloyed with vanadium.* Report IM 1489, Swedish Institute for Metals Research, 1980.

[9] SPEER, J. G., MICHAEL, J. R., HANSEN, S. S. Carbonitride precipitation in niobium/vanadium microalloyed steels. *Metallurgical Transactions A*, 1987, vol. 18A, pp. 211-222.

[10] HILLERT, M., STAFFANSSON, L. I. Regular solution model for stoichiometric phases and ionic melt. *Acta Chemica Scandinavica*, 1970, vol. 24, pp. 3618-3626.

[11] ADRIAN, H. *Numeryczne modelowanie procesów obróbki cieplnej*, Cracow: Wydawnictwa AGH, 2011.

[12] ADRIAN, H., GŁOWACZ, E., AUGUSTYN-PIENIĄŻEK, J., MATUSIEWICZ, P., MARYNOWSKI, P. *Kinetyka wydzielania węglikoazotków w stalach mikrostopowych. W Polska Metalurgia w latach 2011-2014,* Monograph. Cracow : Polski Komitet Metalurgii, 2014, pp. 837-854.

[13] ADRIAN, H., AUGUSTYN-PIENIĄŻEK, J, MARYNOWSKI, P, MATUSIEWICZ ,P. Model kinetyki wydzielania węglikoazotków w stalach mikrostopowych. *Hutnik,* 2014, vol. 81, no. 4, pp. 208-214.

[14] DUTTA, B. Palmiere, E. J., SELLARS, C. M. Modelling the kinetics of strain induced precipitation in Nb microalloyed steels. *Acta Materialia,* 2001, vol. 49, pp. 785-794.

[15] GŁOWACZ, E. *Model kinetyki wydzielania węglikoazotków w stalach mikrostopowych o różnej zawartości azotu.* PhD thesis. Cracow, AGH, 2013.

[16] MAUGIS, P., GOUNE, M. Kinetics of vanadium carbonitride precipitation in steel: A computer model. *Acta Materialia,* 2005. vol. 53, pp. 3359-3367.

[17] PEREZ, M., DUMONT, M., ACEVEDO-REYES, D. Implementation of classical nucleation and growth theories for precipitation. *Acta Materialia.* 2008, vol. 56, pp. 2119-2132.

[18] RAABE, D. *Computational Materials Science*, Wiley-VCH Verlag GmbH, 1998.

[19] HOJNY, M., GŁOWACKI, M. Computer modelling of deformation of steel samples with mushy zone. *Steel Research International,* 2008, vol. 79, no. 11, pp. 868-874.