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Cellular Automata Application for Simulation of Uranium Crystallization Process

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The paper addresses the dynamic modelling of the UNH crystal growth by means of cellular automata(CA) method for nuclear fuel reprocessing as the alternative to traditionally used extraction method. Model-based CA approach is considered to predict dependence of product's properties on input changes: uranium and nitric acid concentration, solution temperature. CA approach is preferable for crystallization process to get a high purity of reprocessed materials. C++ application is developed as a result of the research. The application allows monitoring of dynamics changes of solution temperature, uranium concentration and shape of crystals.

1. Introduction

Advantages of nuclear energy compared to other alternative energy sources and fossil fuels lead to increase of its share in electricity generation. However, problem of spent nuclear fuel handling restrain growth of nuclear share in world's energetics. Current system or nuclear fuel cycle lead to unacceptable growth of spent nuclear fuel handling cost. Crystallization process is considered in this paper as a complement or an alternative to traditionally applied extraction process for high material purification purpose. Crystallization application provide reduction of organic substances and liquid radioactive wastes volume during fuel reprocessing (Veselov et al., 2015). As a result, crystallization allows getting desired purity of output materials. Crystals purity depends on accuracy of control of the process. In seeded cooling crystallization, that is researched in this paper, supersaturation is generated by external cooling of crystallizer. High purity of reprocessing is achieved holding supersaturation on desirable level that corresponds with growth of crystals and non-appearance of new undesirable seeds. Using cellular automata it is possible to monitor the structure of crystals and change adjustments of crystallization process keeping desirable purity of output material up.

2. Cellular automata for crystallization process

Cellular automata are discrete dynamic models whose behaviour is completely specified in a local dependence terms. Cellular automata may be considered as a space divided with a regular grid into cells that keep some information about their state (Toffoli, 1990). Time is discrete and set with a number of iterations. A new state of each cell is determined from the current state of the cell and states of cells in its neighbourhood according to the fixed rules of evolution. CA are successfully applied as the alternative to Partial Differential Equations (PDEs) when their solving requires extremely high computational power (Prieto and Gonçalves, 2014).
CA crystallization process description include next steps:
1) determination of rules of the system evolution;
2) determination of processes which occur during crystallization such as latent heat release during crystallization and the decrease of solution concentration in neighbourhood of the crystallized cell;

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3) acceptance of assumptions that simplify implementation of the model but do not affect results of the research.

Hexagonal grid is used in the model and each cell of this grid determines the unit of nitrate solution of uranyl nitrate. The cause of hexagonal grid use is its high isotropy compared with triangular and rectangular grids (Figure 1.a). Hexagonal grid is transformed (Figure 1.b) for its implementation.

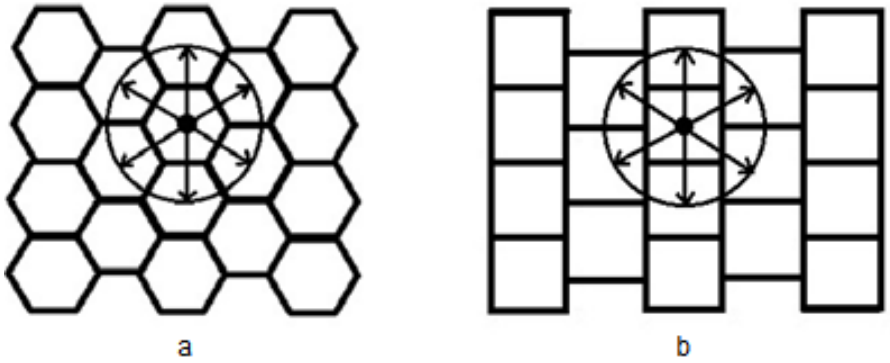

Figure 1: A - hexagonal grid, b - transformed hexagonal grid

Abasheva (2007) has offered a method of CA crystallization modelling: cells of CA have two possible states $\Phi_{i,j}^{(n)} = \{0,1\}$ where 0 corresponds to a liquid state, 1 – solid state, index n determines a number of iteration (time) and indices i,j specify coordinates of cells in the grid. Each cell contain information about its state – concentration $C_{i,j}^{(n)}$ and temperature $T_{i,j}^{(n)}$ – whose initial values are set before simulation. The process of crystal growth is going on under certain rules and conditions of evolution.

Conditions of cell's state transition from a liquid to a solid state:

1) Cell changes its state only from liquid to solid one, there is no reverse transition;
2) Cell is able to crystallize if only there is a solid cell in its neighbourhood;
3) Cell changes its state from liquid to solid one if following condition is satisfied:

$$\Delta_{i,j}^{(n)} > \Delta^* - \lambda \cdot \left[\sum_{k,l \in O(i,j)} \Phi_{k,l}^{(n)} - 3 \right], \quad (1)$$

where $\Delta_{i,j}^{(n)}$ – supersaturation of {i,j}-cell;

Δ^* – limit of supersaturation;

$\Phi_{k,l}^{(n)}$ – state of {k,l}-cells of {i,j}-cell neighbourhood;

λ – a weight coefficient that specify influence of local curvature of interstate surface to the state transition.

State transition of {i,j}-cell from liquid to solid state processes with a latent heat release (dT) of the cell and liquid neighbours concentration decrease $\Delta_{k,l}^{(n)}$:

$$T_{i,j}^{(n+1)} = T_{i,j}^{(n)} + dT, \quad (2)$$

$$C_{k,l}^{(n+1)} = C_{k,l}^{(n)} - q \cdot \Delta_{k,l}^{(n)}, \quad (3)$$

where q – coefficient, that is an analog to kinetic constant of crystal growth rate.

Following equations of heat transfer and diffusion are extremely important for crystallization process:

$$C_{i,j}^{(n+1)} = C_{i,j}^{(n)} + \frac{D}{m}\left(\left\langle C_{k,l}^{(n)} \right\rangle - C_{i,j}^{(n)} \right), \quad (4)$$

$$T_{i,j}^{(n+1)} = T_{i,j}^{(n)} + \frac{a}{p}\left(\left\langle T_{k,l}^{(n)} \right\rangle - T_{i,j}^{(n)} \right), \quad (5)$$

where

$$\left\langle C_{k,l}^{(n)} \right\rangle = \frac{1}{n_l} \sum_{k,l \in O(i,j)} C_{k,l}^{(n)}, \tag{6}$$

$$\left\langle T_{k,l}^{(n)} \right\rangle = \frac{1}{6} \sum_{k,l \in O(i,j)} T_{k,l}^{(n)}, \tag{7}$$

n_l is a number of liquid cells in neighbourhood of the {i,j}-cell ($n_l \le 6$); $\left\langle C_{k,l}^{(n)} \right\rangle$ and $\left\langle T_{k,l}^{(n)} \right\rangle$ – average concentration of liquid cells and average temperature of all neighbours of the {i,j}-cell; D, a, m, p – parameters, that depend on diffusion coefficient, thermal conductivity and space-time discretization – Eq.(6).
Saturation of uranium solution is specified by the following function of temperature and nitric acid concentration obtained by Chikazawa et al (2012).

$$C_U = 10^{\alpha}, \tag{8}$$

where

$$\alpha = 5{,}504 - 1{,}411 \cdot C_H + 0{,}0263 \cdot {C_H}^2 - 0{,}913 \cdot 10^{-2} \cdot T_{abs} + 0{,}088 \cdot 10^{-4} \cdot {T_{abs}}^2 + 3{,}586 \cdot 10^{-3} \cdot C_H T_{abs}. \tag{9}$$

Tabs – solution temperature (K), C_H and C_U – concentrations of nitric acid (mol\L) and uranium (g\L). Eq(8) is obtained from empirical solubility graphs (Figure 2).

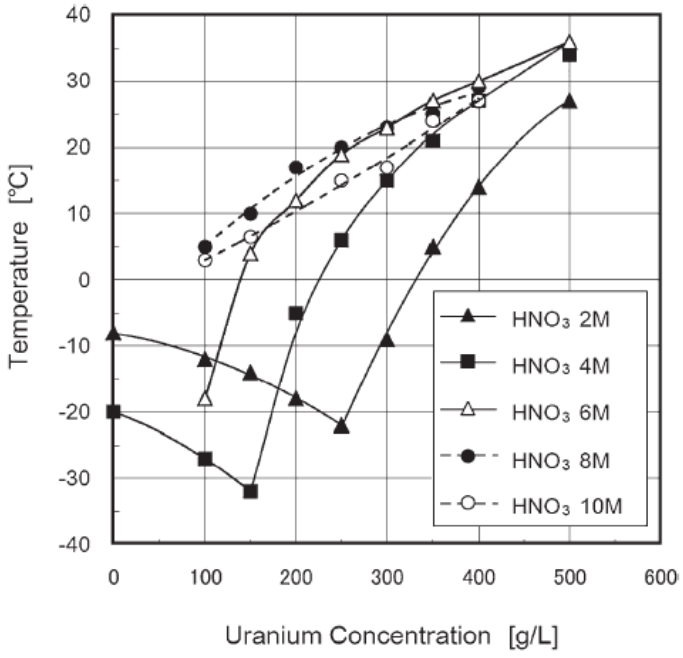

Figure 2: Uranium solubility graphs

3. Results

Next figures represent results of CA simulation of crystallization process. Developed application provides user interface for crystallization process adjusting and displays results of simulation: crystal shape, concentration distribution and temperature distribution (Figure 3). Figure 4 represents crystal growth dynamics for n=10, 20, 30 iterations.

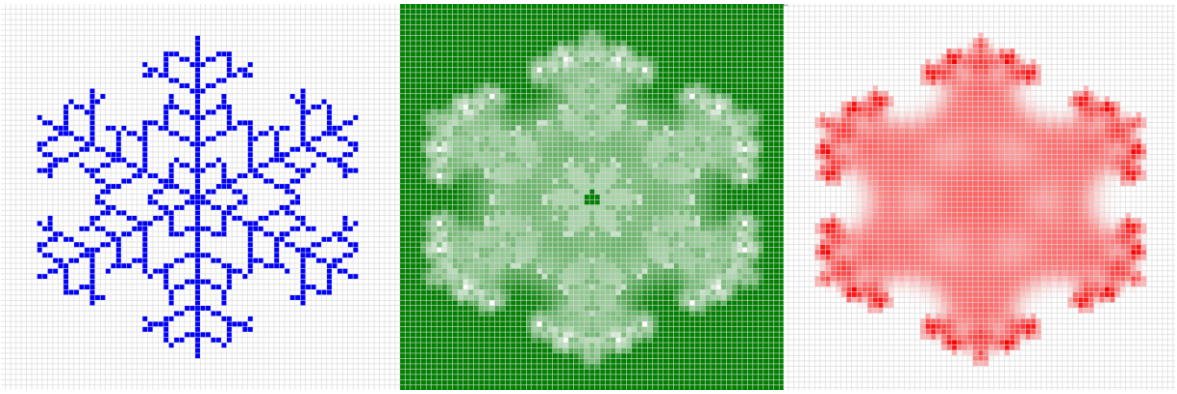

Figure 3: Crystal shape, concentration distribution and temperature distribution

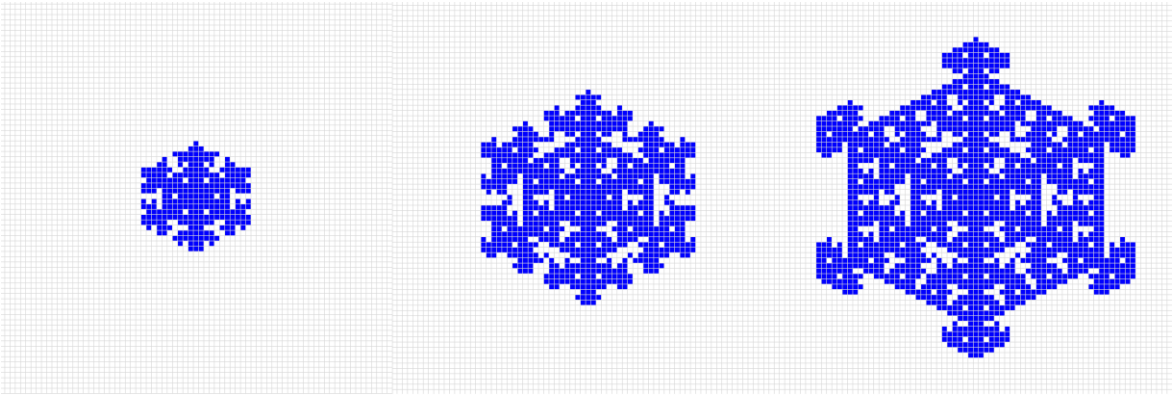

Figure 4: Crystal growth dynamics (n = 10, 20, 30 iterations)

Initial conditions influence analysis and varying of these conditions allow searching for crystallization optimal parameters that provide desirable crystal purity (Figures 5 and 6).

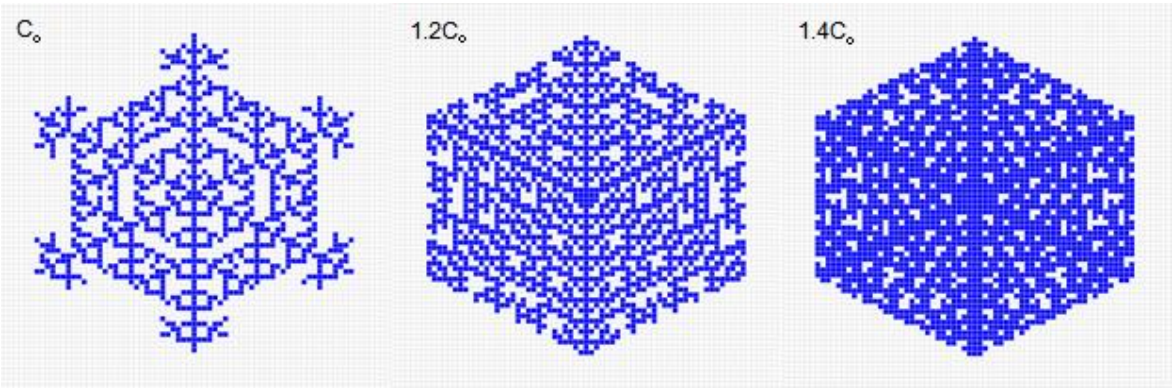

Figure 5: Initial conditions influence (solution concentration increase)

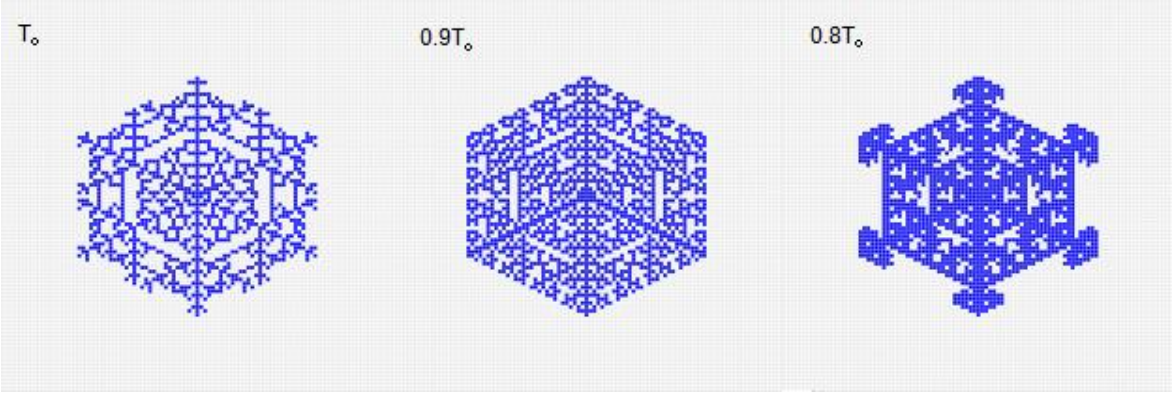

Figure 6: Initial conditions influence (solution temperature decrease)

High influence of diffusion and heat transfer are obvious from Figures 7 and 8. Thus, it is necessary to choose values of their coefficients with a high accuracy.

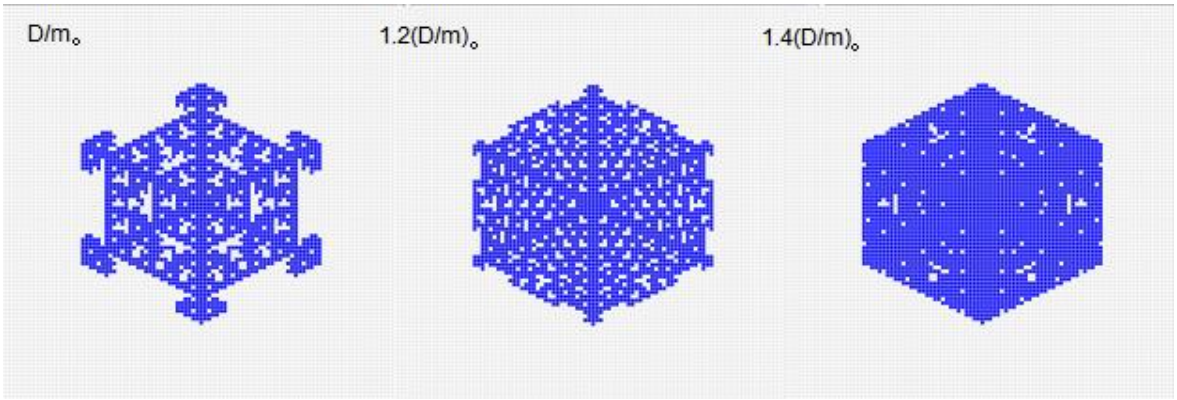

Figure 7: Diffusion influence (diffusion coefficient increase)

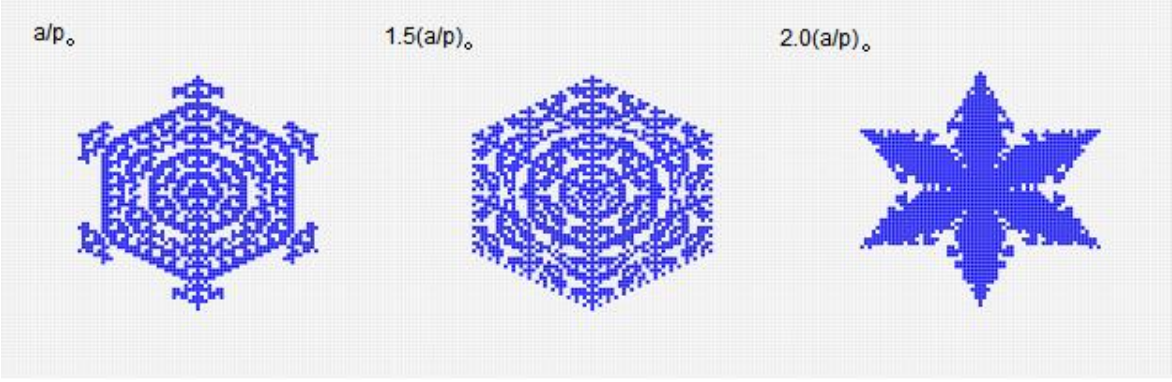

Figure 8: Heat transfer influence (heat transfer coefficient increase)

Table 1 demonstrates dependences of crystal purity on crystallization parameters variation. Purity is calculated as the ratio of solid cells of the crystal to all cells.

Table 1: Purity dependence

Concentration		Temperature		Diffusion coefficient		Heat transfer coefficient	
Variation	Purity	Variation	Purity	Variation	Purity	Variation	Purity
C_0	0.4382	T_0	0.4476	$(D/m)_0$	0.7723	$(a/p)_0$	0.6465
$1.2C_0$	0.6171	$0.9T_0$	0.5905	$1.2(D/m)_0$	0.8169	$1.5(a/p)_0$	0.6877
$1.4C_0$	0.8606	$0.8T_0$	0.7923	$1.4(D/m)_0$	0.9576	$2.0(a/p)_0$	0.9030

4. Conclusion

Application for crystallization process simulation is developed. Presented application allows monitoring of crystallization process dynamics and obtaining information about crystallization output material. Time of simulation is specified with a number of iterations and depends on chosen time-space discretization. Importance of diffusion and heat transfer coefficient's values determination for crystals purity is demonstrated. Crystallization dynamics may be controlled via initial uranium concentration and solution temperature. Uranium saturation for crystallization conditions is calculated as a function of nitric acid concentration and solution temperature.

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