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NUMERICAL INVESTIGATION OF THE INFLUENCE OF PULSED LASER DEPOSITED TiN THIN FILM MORPHOLOGY ON DEFORMATION INHOMOGENEITIES

Investigation of influence of TiN thin film morphology on deformation inhomogeneities is an overall subject of the research. Numerical modelling approach that was selected for the study is based on the digital material representation concept, which gives an opportunity to directly replicate columnar microstructure morphology of an investigated thin film. Particular attention in this paper is put on the discussion of the influence of cellular automata neighbourhood on thin-film digital morphologies and their further deformation behaviour. Additionally, an evaluation of representativeness aspects of the digital models, in particular, the analysis of the influence of a number of columns, their dimensions and variations in their properties on the material behaviour during compression tests is also presented. The non-periodic boundary conditions are assumed during the investigation. Obtained data in the form of equivalent stress distributions as well as homogenized stress-strain curves from analyzed case studies are presented and discussed within the paper.

Keywords: thin films, pulsed laser deposition, digital material representation, homogenization, finite element method

1. Introduction

Hybrid products, in which the base material is covered by a thin layer of another material are very attractive for the market as they give a possibility to provide desired, specific in-use properties [1]. One of the methods to obtain such coatings in the form of thin films is a deposition process by a laser ablation [2,3]. Deposition processes are especially interesting in the bioscience applications providing products with required strength, optical and bio-protection properties [2,4]. However, in these applications, any kind of internal defect within the deposited thin-film structure is unacceptable as it may lead to local stress and strain localisation during further exploitation. Such defects can also cause a build-up of biological material or be responsible for the deterioration in strength properties, which may be hazardous for a patient. At the same time, selection of the deposition process parameters directly influences the final thin film surface morphology, which is crucial in the case of e.g. optical properties [5-7].

That is why it is extremely important that the surface, as well as the morphology of deposited thin films, ensure all in-use expectations required for their exploitation conditions. However, the mechanical behaviour of such nanoscale materials often differs from their macroscale counterparts. Therefore, these kinds of thin-film materials have to be subjected to sophisticated plastometric tests e.g. micropillar compression [8-10] or nanoindentation [11] in order to evaluate their behaviour under loading at an appropriate length scale. Interpretation of these experimental results also becomes more elaborated in comparison to standard macroscopic evaluations, and that is why it is often supported by dedicated numerical simulations. However, due to mentioned complex columnar morphology of as-deposited thin films, the numerical analysis of their behaviour under loading conditions with conventional modelling approaches is usually simplified [12-15] and seems to be insufficient [16]. That is why for improving the quality of numerical simulations, especially at the micro/nanoscale level, the exact material morphology has been considered by the authors. For that, the Digital Material Representation Finite Element (DMR-FE) modelling approach was adapted to nanoscale structure morphologies. Authors have already proven good predictive capabilities of this concept in their previous work [17], where different DMR-FE models of TiN/Si thin-films were generated by the cellular automata growth algorithm and their behaviour under deformation conditions was then analysed. It was concluded from that research that the local heterogeneities in stress or strain fields can be found especially

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in the TiN/Si interface area and along columns boundaries. Therefore, in the current study influence of columns shapes, their number and variations in the hardening properties within the DMR-FE models was addressed to extend the knowledge on their predictive capabilities.

The summary of the previously developed cellular automata (CA) – Monte Carlo (MC) column growth algorithm is presented in the first part of the paper. Then, a numerical compression test of the TiN/Si(100) films with non-periodic boundary conditions is investigated as a case study. Such a model is the basis for evaluation of a representativeness aspect of the digital morphology models, in particular, mentioned analysis of an influence of CA neighbourhood type, number of columns, their shapes, and finally, variation in properties on a thin-film response during deformation.

2. Digital Material Representation of TiN/Si(100) thin films

The pulsed laser deposition (PLD) is one of the laser ablation methods that can provide a wide range of thin films [1]. In the PLD a high-power laser beam is focused periodically onto a target material provoking instantaneous evaporation and ionization of surface atoms. This source forms a plasma inside the plume of the evaporated material. Atoms, electrons and ions are driven away from the target at high speeds into the vacuum of controlled conditions. High-speed strike onto the surface of a substrate leads to nucleation and growth of thin-films with the same chemical composition as an evaporated material. Gas molecules can be additionally introduced inside the reaction chamber in order to react with the atoms or molecules from the plume allowing the deposition of a thin film on a substrate surface [18,19]. Depending on the deposition parameters, different thin-films structures can be obtained. The most important parameter controlling the layer morphology is the homologous temperature, which can be described as the ratio of the substrate temperature to the melting point (T_S/T_m). A lot of studies on relations between the mentioned temperature and final thin-film morphologies can be found in the scientific literature [20,21]. The most frequently used model describing deposited morphologies is based on the Thornton [21] work. The Thornton model takes into account an influence of the homologous temperature during deposition on a final columnar morphology as seen in Fig. 1.

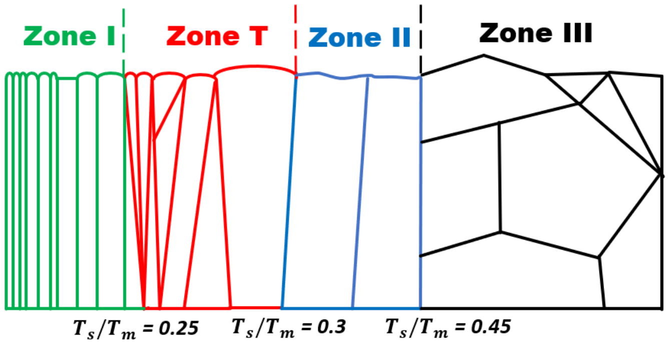

Fig. 1. The concept of the Thornton deposition model [21,22]

Because TiN/Si thin-films grow according to the Zone 1 in the Thornton model, all DMRs generated within this research should consist of columns arranged side by side without significant changes in the width measured from the substrate to the surface of the thin layer. Different types of cellular automata neighbourhoods [23,24] can be used to generate the thin-film digital models [17,25]. This may, however, have an impact on the final morphology, especially the shape of the borders between columns. To investigate this issue, three commonly used CA neighbourhoods: Moore, hexagonal random, and pentagonal random have been used (Fig. 2).

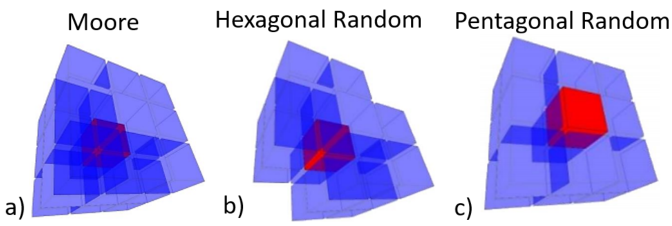

Fig. 2. a) Moore, b) hexagonal random and c) pentagonal random neighbourhoods of the CA cell marked in red

To clearly point out the possible influence of these three CA neighbourhoods on final columns morphology, the single-column was generated as a case study (Fig. 3).

As seen in Fig. 3, due to the random character of the CA neighbourhoods, both hexagonal and pentagonal neighbourhoods result in column roughness. Apparently, this is more pronounced in the case of pentagonal random neighbourhood, as presented in Fig. 3c.

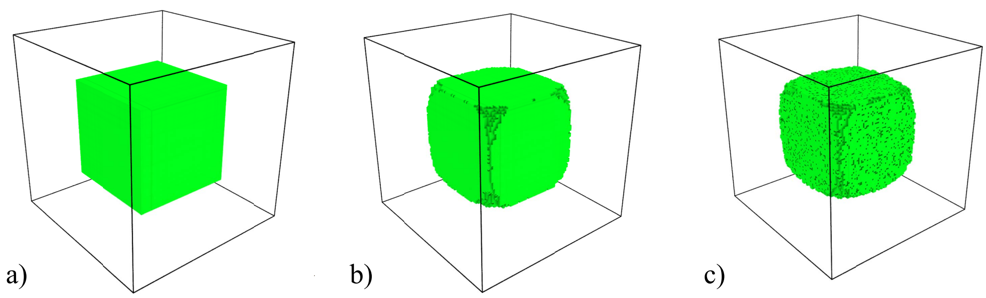

Fig. 3. Three dimensional models of a single column generated with the CA growth model and different neighbourhood types a) Moore, b) hexagonal random and c) pentagonal random

To investigate this issue in a more practical model of a columnar thin-film morphology, a set of fifteen different DMR models with an increasing number of columns from 25 to 400 was generated. Again, three mentioned CA neighbourhoods were used during the model's generation stage. Height of columns was selected as 180 nm while substrate thickness as 20nm to replicate metallographic observations from earlier authors works [17,26]. All generated DMRs are presented in Fig. 4-6.

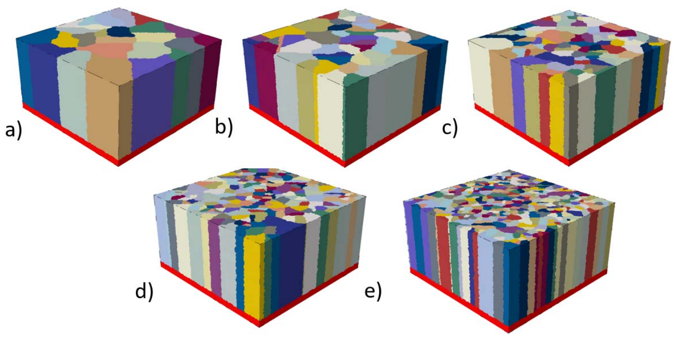

Fig. 4. Generated, with the Moore CA neighbourhood, set of digital models of TiN thin-films with a) 25 b) 50 c) 100 d) 200 and e) 400 columns

Influence of CA neighbourhoods on thin-films morphologies was quantified by evaluation of contact area sizes between neighbouring columns. It was assumed that the increase in surface roughness of each column after the growth stage will indicate the CA neighbourhood influence. Therefore, Fig. 7-9 presents an example of such a comparison for 25 and 400 columns obtained with three investigated CA neighbourhoods.

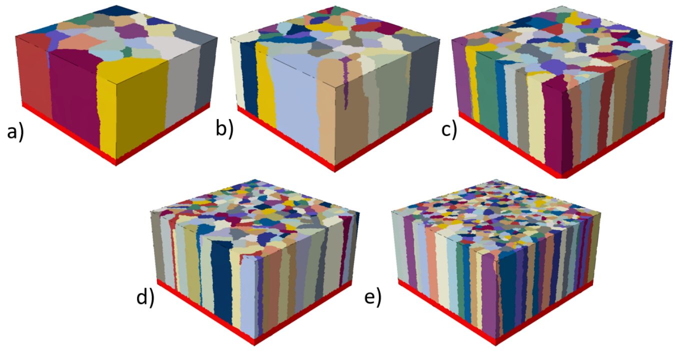

Fig. 5. Generated, with the hexagonal random CA neighbourhood, set of digital models of TiN thin-films with a) 25 b) 50 c) 100 d) 200 and e) 400 columns

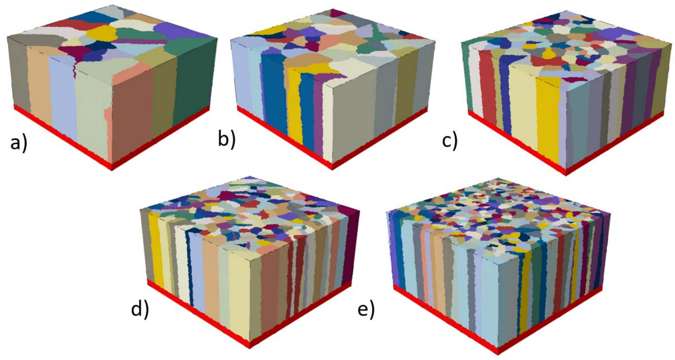

Fig. 6. Generated, with the pentagonal random CA neighbourhood, set of digital models of TiN thin-films with a) 25 b) 50 c) 100 d) 200 and e) 400 columns

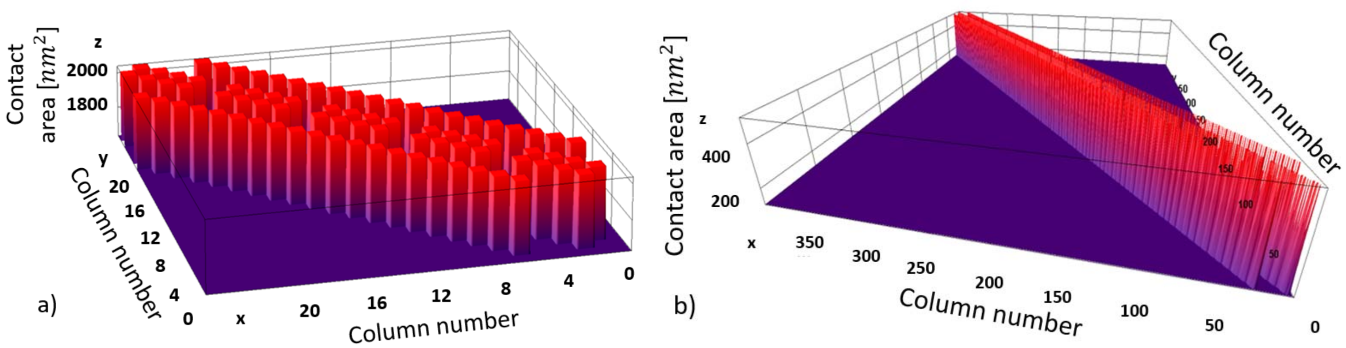

Fig. 7. Contact area between columns obtained with the Moore CA neighbourhood for a) 25 b) 400 columns

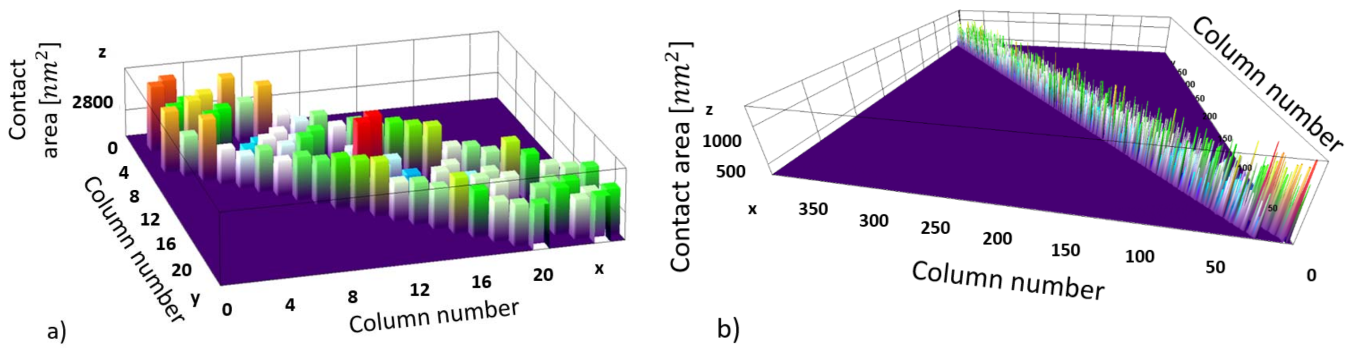

Fig. 8. Contact area between columns obtained with the hexagonal random CA neighbourhood for a) 25 b) 400 columns

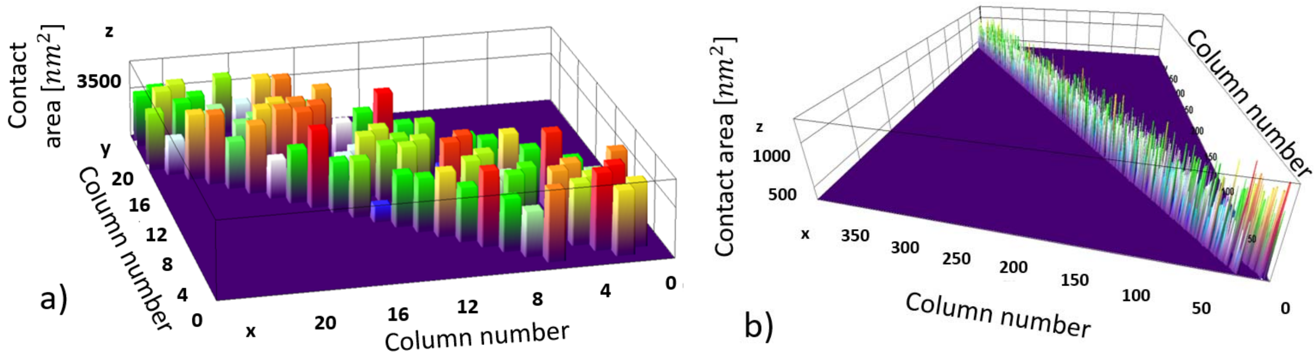

Fig. 9. Contact area between columns obtained with the pentagonal random CA neighbourhood for a) 25 b) 400 columns

As presented the highest roughness between columns is within the models generated with the pentagonal random neighbourhood. The hexagonal random neighbourhood also results in significant variation in contact surface area. The most uniform geometry of columns is obtained for the Moore neighbourhood, which is in agreement with the results obtained with the simple case study from Fig. 3. Therefore, to analyse if these variations influence DMR models behaviour under loading conditions, all previously presented thin-films morphologies were discretized with the *DMRmesh* software [27] and used as input for the finite element calculations. In this case, the finite elements have to be highly refined along the boundaries of the columns to properly capture their geometries and to further capture the solution gradients that are expected in these regions due to differences in hardening behaviour of subsequent columns. The number of finite elements ranges from approx. one million to four million. Example of the generated and discretized DMR model with 25 columns is presented in Fig. 10.

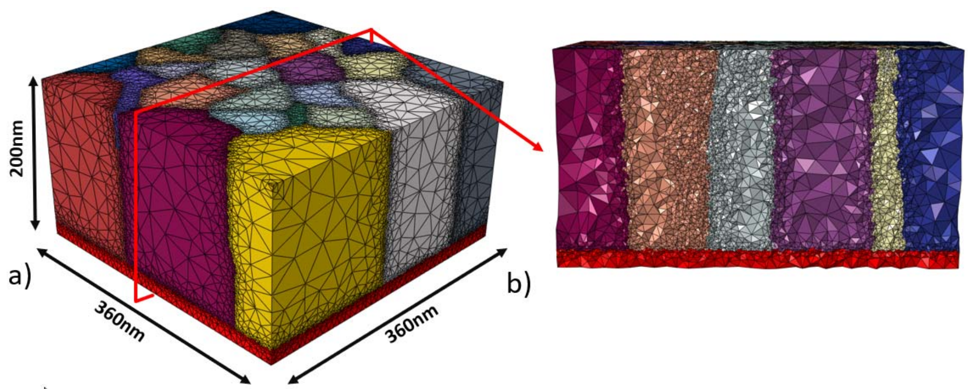

Fig. 10. Example of a 3D DMR model of a thin-film generated with the hexagonal random neighbourhood a) full model and b) cross-section

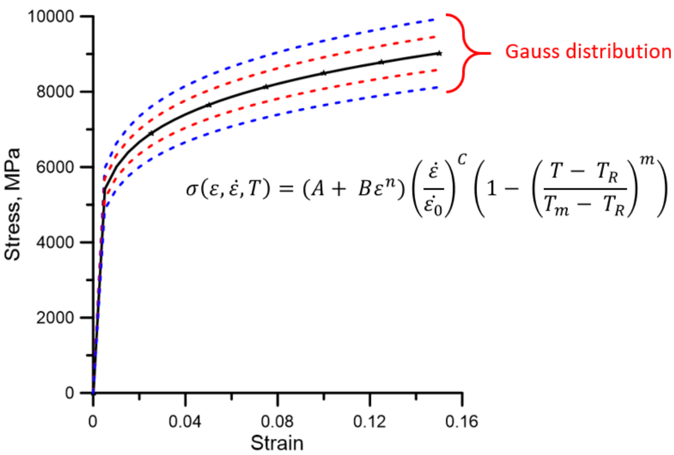

Fig. 11. Schematic representation of flow stress curves generated with the Gaussian distribution function (where: A, B, C, n, m – material constants, σ – plastic flow stress, ε – equivalent plastic strain, $\dot{\varepsilon}$ – equivalent plastic strain rate, $\dot{\varepsilon}_0$ – reference equivalent plastic strain rate, T_m – melting temperature, T_R – transition temperature)

The possibility to assign material properties to the Si substrate and particular TiN columns is the main advantage of the developed DMR-based numerical model. Elasto-plastic material properties of Si and TiN thin-films were obtained from the nanoindentation test and inverse analysis as described in earlier work [26]. To differentiate the hardening behaviour of subsequent columns a slightly different flow stress model parameters were assigned. There are many examples in the literature where thin films are analyzed for differentiation in stress distributions resulting from their structure [28,29]. In order allow the analysis of the influence of the size and morphology of the columns and their crystallographic orientations on the mechanical response of the TiN thin-films, the simplification of modelling with the use of Gauss decomposition was used. In this work, the random Gauss distribution with three standard deviations 5%, 25% and 50% of the B parameter (Table 1) in the Johnson-Cook flow stress model was used (Fig. 11). The higher the standard deviation the more heterogeneous behaviour of columns within the thin-film is expected.

TABLE 1

Johnson-Cook model parameters for titanium nitride

Material	*A* [MPa]	*B* [MPa]	*n*	*m*	*C*	$\dot{\varepsilon}_0$	T_m [°C]	T_R [°C]
TiN	2000	7000	0.05	0.3	0.022	0.01	3000	20

3. Numerical investigation of non-periodic DMR of thin film

All 45 generated models (three sets of five models in combination with three different spreads of properties) were subjected to numerical simulation of the compression test. This test was selected to evaluate thin-film responses to loading. Models were developed in the commercial ABAQUS application. The upper tool was assumed to be rigid during the analysis. The sample bottom surface nodes were fixed in *z*-direction, while symmetric boundary conditions were applied to the side surface nodes. The model assembly with applied boundary conditions is shown in Fig. 12.

As seen in Fig. 12, the upper tool is displaced towards the sample by 100 nm ($\varepsilon_{eng} = 0.5$). Examples of obtained equivalent stress distributions after the compression are presented in Fig. 13.

As seen in Fig. 13, an increasing number of columns results in an increased number of locations where stress inhomogeneities can be observed. It is also clearly visible that the stress variations are between columns within the thin film. These zones where differences between stresses are the highest are usually a precursor of material failure between subsequent columns.

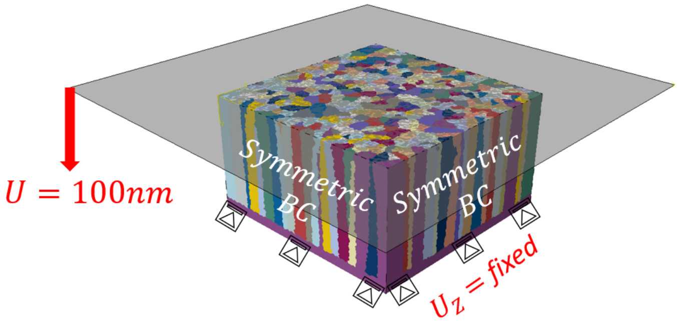

Fig. 12. Assembly of the compression test with applied boundary conditions

Additionally, the homogenization procedure was performed in order to get quantitative information on stress-strain relationships in thin-film models [30]. Homogenised stress-strain curves calculated from all 45 models are presented in Fig. 14.

As seen in Fig. 14, obtained differences are quite small especially for the Gauss spread approx. 5%. That is why a more detailed investigation based on curves integrals was also performed, as presented in Fig. 15.

As seen in Fig. 14a-c obtained differences in flow stress values are not significant throughout the deformation. For all models from 25 to 400 columns curves are very similar with fluctuations less than 1% to each other. But when Gauss spread is 25% this fluctuation is different especially for the models with 25 and 50 columns (Fig. 14d-f). The integral value from the chart is different between curves up to 9%. In the models containing a larger number of columns, this area disproportion decreases

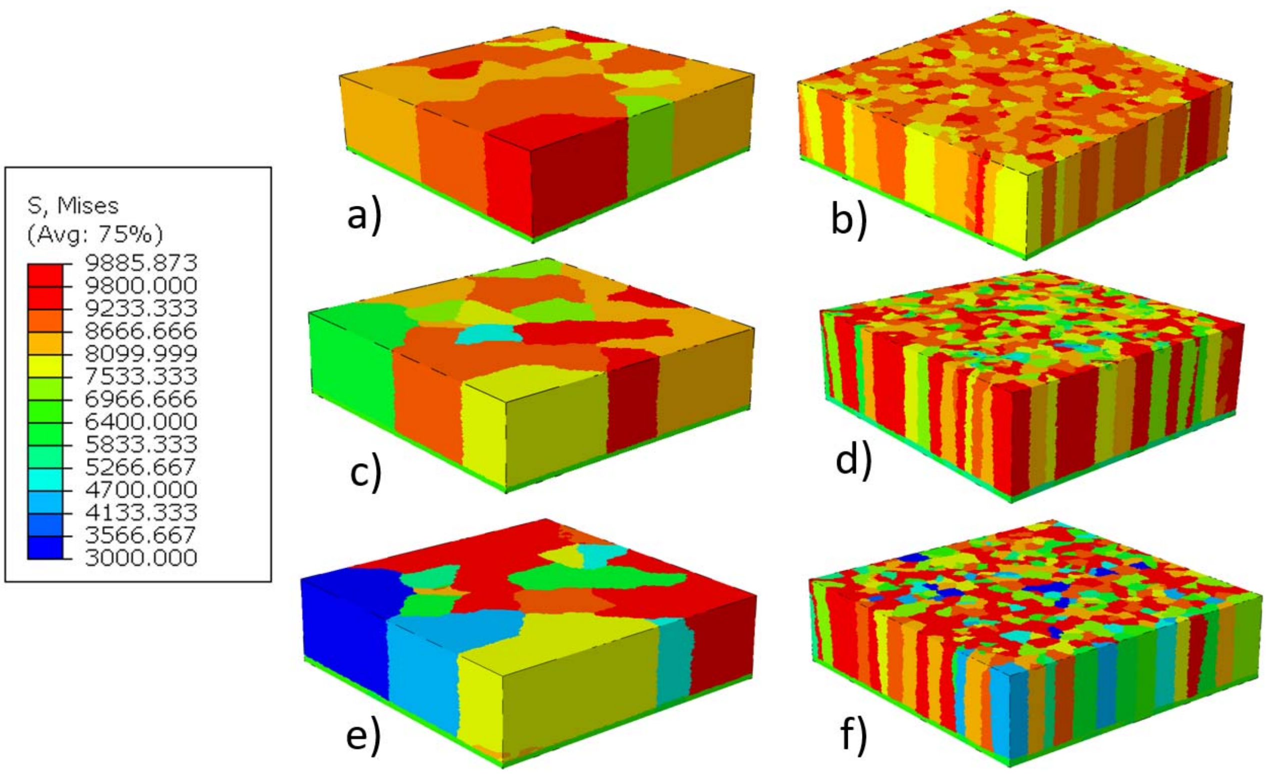

Fig. 13. Example of the stress distribution in the thin films created with hexagonal CA neighbourhood 25 columns and 400 columns for Gauss distribution spread a,b) 5% c,d) 25% and e,f) 50%

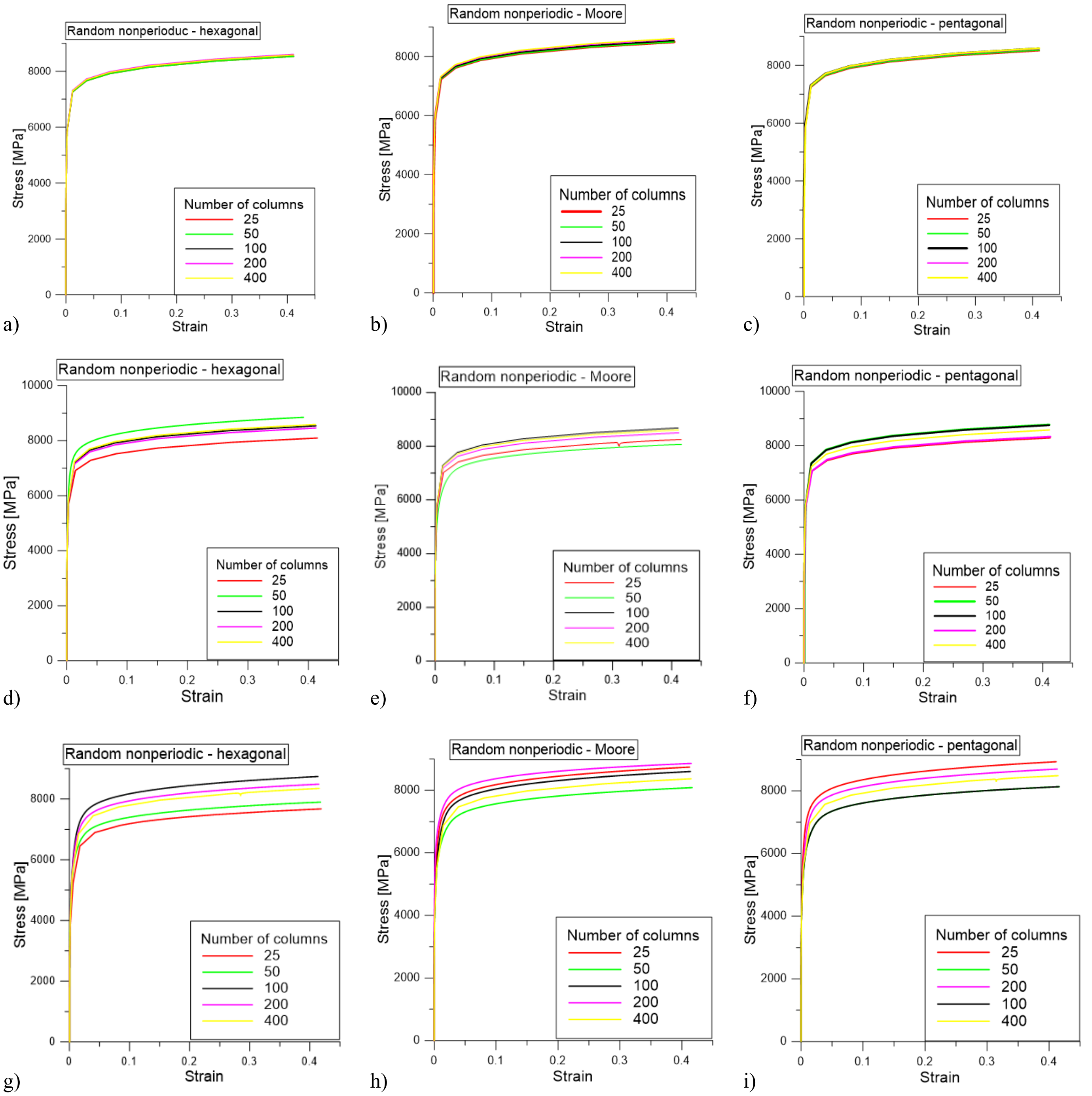

Fig. 14. Homogenized stress-strain curves after deformation of thin films with different numbers of columns and Gauss spread of approx. a-c) 5%, d-f) 25% and g-i) 50%

what can be seen on the homogenised curves. But when Gauss spread is 50% then fluctuations are much larger and the results are significantly different (Fig. 14g-i). In this case, it can be concluded that the size of the selected DMR model is not representative when the material properties of the individual columns are characterized by large differences. For a large difference in material properties in thin-films, a larger DMR should be used.

Another observation is related with influence of the CA neighbourhoods on the thin-film mechanical response. It can be seen that the neighbourhood choice has a small or minimal influence on the obtained results. Previously observed large roughness between columns generated with the hexagonal and pentagonal random neighbourhoods are now imperceptible. The reason for such behaviour may be caused by the mesh discretization algorithm, which additionally smooths out the discretized column surfaces. This can be again investigated in a simple case scenario from Fig. 3. Single columns after the discretization process are presented in Fig. 16.

As seen in Fig. 16 after the finite element mesh generation differences between surface shapes are minimal, eliminating any possible influence of the CA neighbourhood type on obtained results.

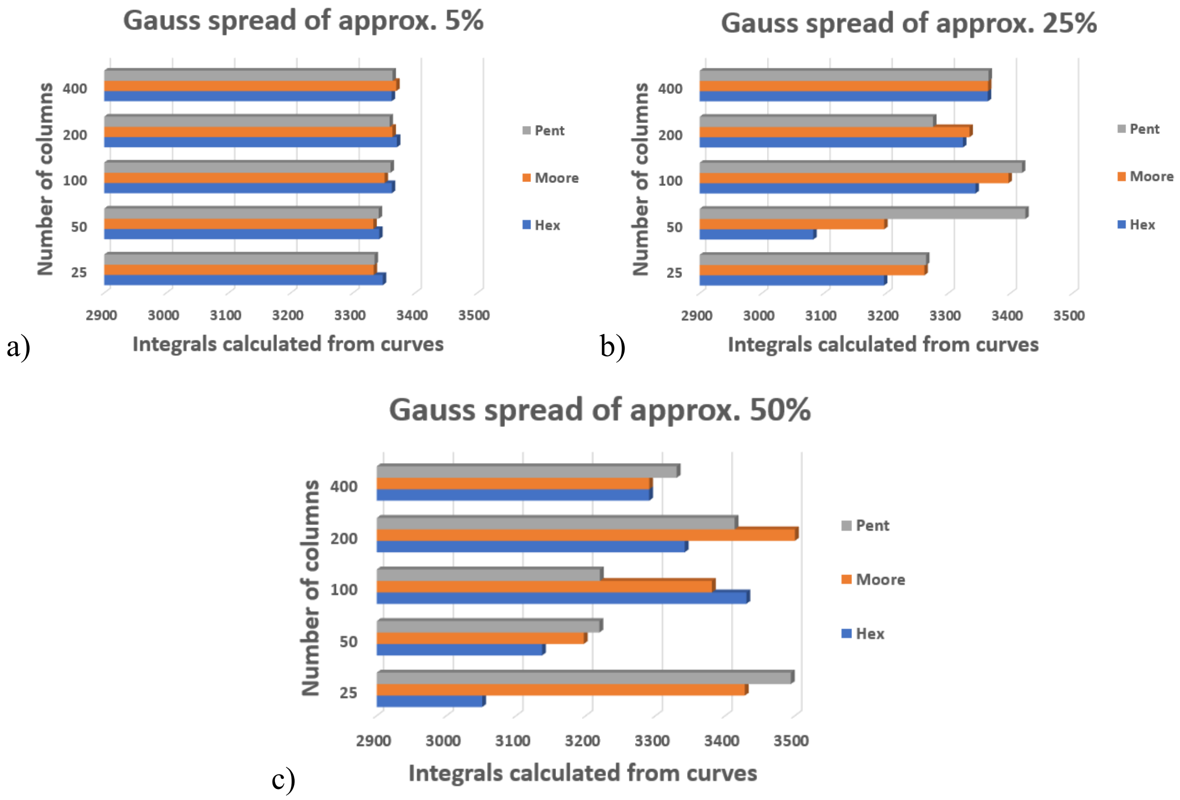

Fig. 15. Integrals calculated from stress-strain curves after homogenisation procedure for different Gauss spreads of approx. a) 5%, b) 25% and c) 50%

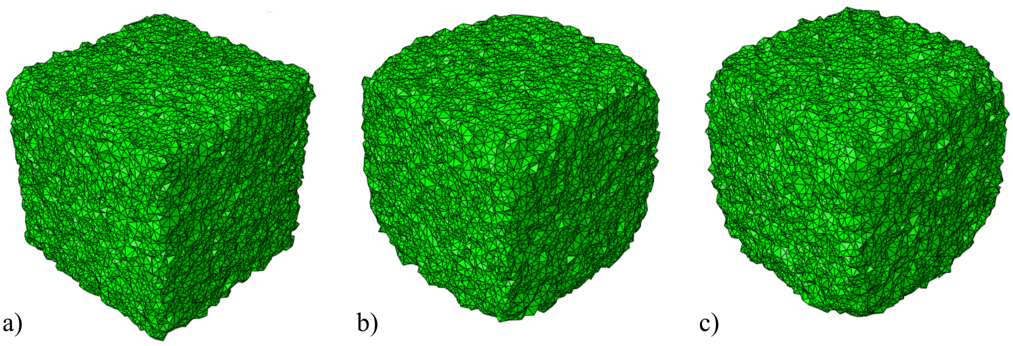

Fig. 16. Models from Fig. 3 generated with a) Moore, b) hexagonal random and c) pentagonal random neighbourhoods after finite element mesh discretization

As presented throughout the paper, the fundamental study on the DMR model representativeness aspects, in such complex materials like deposited thin films is of importance. Various material related as well as numerical related aspects affect the model response during the calculations. Therefore, such fundamental research is required to ensure the robustness of the digital material representation model of a thin film.

4. Conclusions

Based on the presented results it can be concluded that:

- the digital material representation model of the deposited thin films allows predicting inhomogeneities in stress fields under deformation conditions,
- for the small 5% difference between material properties, there is no influence of the digital material morphology on film behaviour under compression,
- the larger variations in columns properties, the larger DMR size is considered as representative volume element,
- the proposed contact area evaluation procedure seems to be a good indicator for surface roughness between the columns,
- the CA neighbourhood type used during the thin-film model generation has an impact on final column morphology. The random pentagonal neighbourhood provides the highest surface roughness,
- finite element discretization algorithm smooths borders of the columns, which reduces the mentioned impact of the CA neighbourhood type on its behaviour under loading conditions.

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