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INFLUENCE OF THE FORM OF SHORT-RANGE REPULSIVE INTERACTIONS ON FINAL SHAPE OF INTERNAL STRUCTURE IN MAGNETO-RHEOLOGICAL FLUIDS

The presented paper is devoted to the task of computational simulation of ferromagnetic particles in magneto-rheological fluids. Under the action of an external magnetic field, ferromagnetic particles form a complex internal microstructure. This microstructure is generally parallel to the direction of the external magnetic field intensity vector. The main aim of this work is to investigate the influence of the short-range repulsion interactions between the particles on the final shape of the internal microstructure. This interaction is implemented in the simulation process in such a way that the moving particles do not overlap. In the adopted theoretical model, in addition to the effects of short-range repulsion interactions, magnetostatic and hydrodynamic interactions are also taken into account. It is worth stressing that the applied theoretical model is very simple, however, it enables estimation of the mentioned effect. It was assumed that all the ferromagnetic particles have a spherical shape with a constant radius. A series of two-dimensional numerical simulations is carried out based on the molecular dynamic algorithm. The short-range repulsive interactions are described by a polynomial and by an exponential function with various parameter values. It turned out that the final shape of the microstructure strongly depends on the applied form of short-range repulsion. It is possible to obtain single isolated strings of particles as well as complex structures known as particle clusters.

Keywords: magneto-rheological fluids, ferromagnetic particles, computer simulation, molecular dynamics, short-range repulsive interactions, microstructure

WPŁYW KRÓTKOZASIĘGOWYCH ODDZIAŁYWAŃ ODPYCHAJĄCYCH NA KOŃCOWY KSZTAŁT STRUKTURY WEWNĘTRZNEJ DLA PŁYNÓW MAGNETO-REOLOGICZNYCH

Prezentowana praca poświęcona jest zagadnieniu komputerowej symulacji zachowania cząstek ferromagnetycznych w płynach magneto-reologicznych. Pod działaniem zewnętrznego pola magnetycznego wspomniane cząstki tworzą skomplikowaną mikrostrukturę wewnętrzną. Generalnie, mikrostruktura ta wykazuje uporządkowanie w kierunku równoległym do wektora natężenia zewnętrznego pola magnetycznego. Głównym celem pracy jest wykazanie wpływu krótkozasięgowego oddziaływania odpychającego cząstki na ostateczny kształt owej mikrostruktury. Oddziaływanie to jest wprowadzane w symulacji po to, aby pozycje cząstek nie nakładały się wzajemnie. W przyjętym modelu teoretycznym, oprócz oddziaływań krótkozasięgowych, uwzględniono jeszcze oddziaływania magnetostaticzne oraz hydrodynamiczne. Jest to możliwe najprostszemu modelowi cieczy magneto-reologicznej pozwalającemu jednakże zbadać wpływ analizowanego oddziaływania krótkozasięgowego. Założono, że wszystkie cząstki posiadają kształt sferyczny o identycznym promieniu. Przeprowadzono szereg symulacji dwuwymiarowych w oparciu o algorytm dynamiki molekularnej. Oddziaływanie krótkozasięgowe opisano za pomocą wielomianu oraz funkcji wykładniczej. W przypadku funkcji wykładniczej rozważano różne wartości parametrów. Po przeprowadzeniu symulacji uzyskano różne kształty mikrostruktury wewnętrznej, począwszy od pojedynczych izolowanych łańcuchów cząstek po stosunkowo skomplikowane struktury zwane *klastrami* cząstek.

Słowa kluczowe: cieczy magneto-reologiczne, cząstki ferromagnetyczne, dynamika molekularna, krótkozasięgowe oddziaływania odpychające, mikrostruktura

INTRODUCTION

Magneto-rheological (MR) liquids were discovered by Rabinow [1] in 1948. These fluids are a colloidal dispersion of ferromagnetic particles immersed in a magnetically indifferent carrier fluid [2]. When an external field is switched on, the particles become polarized and they start to form chain-like structures, which are generally parallel to the direction of the vec-

tor of the external magnetic field intensity. On the macro scale this process causes a rapid change in the physical and mechanical properties of the MR fluid. The liquid begins to behave like a solid body. Moreover, a clear yield point, whose magnitude depends on the external magnetic field intensity applied, is observed. Thus MR fluids are considered as smart ma-

materials and they are used in modern devices like semi-active dampers [3], clutches and brakes [4] or valves [5]. On the macro scale, the physical and mechanical properties are described by different constitutive models [6] and the Bingham model seems to be the most frequently applied. It should be stressed here that the parameters of macro scale constitutive models have to be evaluated in a relevant experiment. It is very difficult to evaluate the properties of MR fluid theoretically with a sufficiently high accuracy at the design stage. Therefore, the process of manufacturing MR fluids with the required optimal physical and mechanical properties is very difficult and time consuming. This results from the many complicated phenomena occurring in the process of microstructure formation inside the MR fluid under the action of the external magnetic field. Moreover, these phenomena can be investigated only on a micro scale. Therefore, it seems reasonable to adopt the Molecular Dynamic (MD) simulation [7] or Monte Carlo (MC) algorithm [8] in order to simulate the changes in the microstructure of MR fluids.

The MC algorithm was used successfully by Chantrell et al. [9], Satoh [10], Castro et al. [11] for a particle system where the particles have different diameters, or Satoh [12] for rod-like particles. Although the MC approach is much easier in practical use, it merely allows analysis of the final microstructure. It is only known that the obtained pattern of particles provides a minimum of the total potential energy. Other parameters like microstructure formation time, velocity of the moving particles, etc. are not available.

In contrast to the MC method, MD simulations allow one to obtain a complete picture of the phenomena occurring in the MR fluid when the external magnetic field is switched on. Chronologically, one of the first MD simulations was carried out for electrorheological (ER) liquids (Klingenberg et al. [13]). For ER fluids, the fundamentals of MD simulations can be found in the review by Klingenberg and Parthasarathy [14]. Taking into consideration the analogies between electrostatic and magnetostatic phenomena, the presented theoretical model of an ER fluid can be easily adopted for MR fluids. This approach, with various modifications, was used by Ly et al. [15], Murashov and Patey [16], Martin et al. [17], Huang et al. [18] or Li Qiang et al. [19]. However, the real challenge is to simulate the flow of a MR fluid when the external magnetic field is active in order to obtain a change in viscosity or to observe the appearance of the yield point, the so-called MR effect. This problem was studied by Satoh et al. [20, 21] for ferromagnetic fluids, who carried out full three-dimension simulations. The hydrodynamic effects are described with the aid of the modified Stokesian method. In this kind of liquids, the ferromagnetic particles are much smaller in comparison with classical MR fluids. Instead of a yield point, an increase in the dynamic viscosity is observed. The simulation of MR fluid flow through a channel was investigated by Pappas and Klingenberg [22]. It was shown in this work

that in order to obtain a clear yield point, it is necessary to include in the simulation the interaction (friction) between the moving particles and the channel wall. Joung and See [23, 24] performed the simulation of an MR fluid where the effects connected with hydrodynamic interaction were investigated. They used the Rotne-Prager-Yamakawa tensor, which describes these phenomena. The interaction between the channel walls and particles was also studied. The flow of ferromagnetic non-spherical nanoparticles was simulated by Enomoto et al. [25].

To summarize, in order to perform the simulation of a ferromagnetic particle system, at least three main interactions have to be defined, namely: magnetostatic, hydrodynamic and short range repulsive forces. Most frequently, the magnetostatic forces are determined with the aid of a two-body magnetic potential, which describes the energy between two interacting magnetic dipoles [26]. It is worth noting here that Ly et al. [15] used the modified boundary element method (Fast Multiple Method) in order to evaluate the internal magnetic field distribution inside the simulation area. Nevertheless, this method is very time consuming and difficult in application. Depending on the aim of the simulation, the hydrodynamic interactions can be described with the use of complicated relationships or by a simple, linear equation [14]. The last interaction is introduced in order to prevent the moving particles from overlapping during the simulation. Sometimes it can be interpreted as the force caused by the special surfactant layer that covers the ferromagnetic particles [20]. This interaction can be defined with the use of a different equation, i.e. polynomial, exponential or logarithmic [14]. In the opinion of the author of the present work, this interaction could have a significant impact on the final shape of the particle aggregations obtained as a result of the MD simulation. Depending on the form and parameters that describe the short-range repulsion in question, the obtained clusters of ferromagnetic particles can be quite different. To the author's knowledge, there are no publications where this problem is investigated, which is the main motivation behind the currently presented analysis.

THEORETICAL MODEL OF MR FLUID

It is assumed that the investigated MR fluid consists of N spherical particles made of an identical ferromagnetic material in which radius a of all the particles is constant. The particles are immersed in a magnetically indifferent carrier fluid with dynamic viscosity η . The particles can move only in two directions (2D simulation). The area of simulation is a square with edge length l_s . Periodic boundary conditions [7] are assumed. This means that the simulation area is surrounded by squares where the particle distribution is the same at each time step. The idea of periodic boundary conditions with a cut-off radius is shown in Figure 1. If the

particles move out of the simulation area, other particles should be added on the opposite side. In the case of fluid flow simulation, the Less-Edwards boundary condition can also be applied [27].

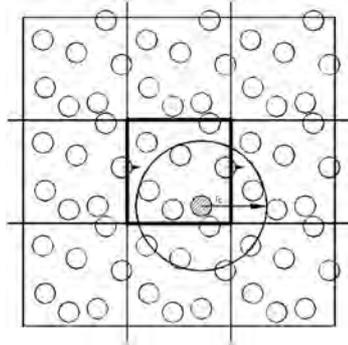


Fig. 1. Periodic boundary condition [7]

Rys. 1. Okresowe warunki brzegowe [7]

The volume fraction is calculated in the following way. Because the particles are spherical in shape, the volume, which is filled with the carrier fluid, is a rectangular prism with dimensions $l_S \times l_S \times 2a$. Finally, the volume fraction is determined with the use of the expression:

$$v_f = \frac{2\pi a^2}{3l_S^2} N. \quad (1)$$

In order to evaluate the net force that acts on each particle, three main phenomena are taken into account: magnetostatic interactions, hydrodynamic resistance and short-range repulsion.

Magnetostatic interactions

The force acting between a pair of constant magnetic dipoles, m_i and m_j , can be derived from the magnetostatic potential energy [26]:

$$U_{ij}^m(r_{ij}) = \frac{1}{4\pi\mu_0} \left[\frac{m_i \cdot m_j}{|r_{ij}|^3} - 3 \frac{(m_i \cdot r_{ij})(m_j \cdot r_{ij})}{|r_{ij}|^5} \right], \quad (2)$$

where r_{ij} is the vector which links the geometrical centers of the interacting particles $r_{ij} = r_i - r_j$ and μ_0 denotes the magnetic permeability of the vacuum. The magnetic dipoles can be calculated as follows [28]:

$$m_k = \mu_0 \frac{4}{3} \pi a^3 \chi^{\text{eff}} H_0, \quad \chi^{\text{eff}} = \frac{3\mu_p}{\mu_p - 3\mu_f} \quad k = i, j, \quad (3)$$

where μ_p , μ_f are respectively the relative magnetic permeability of the particles and carrier fluid, H_0 is the vector which describes the external magnetic field intensity. Nevertheless, in the case of real MR fluids, effective relative magnetic susceptibility χ^{eff} is a function of the volume fraction. Thus the effective value of χ^{eff} in Eq. (3) can be evaluated alternatively as:

$$\chi^{\text{eff}} = \mu^{\text{eff}} - 1, \quad (4)$$

where μ^{eff} is the effective relative magnetic permeability and its values can be found in [29] and they are as follows: $\mu^{\text{eff}}(v_f = 10\%) = 3.07$, $\mu^{\text{eff}}(v_f = 20\%) = 5.02$, $\mu^{\text{eff}}(v_f = 30\%) = 6.97$. The presented relationship has an almost linear character, thus by using simple linear interpolation, the value of μ^{eff} can be easily evaluated for any value of volume fraction v_f . Moreover, it is assumed that the intensity of the external magnetic field is relatively small and it does not induce the saturation phenomenon. Consequently, the linear relationship, Eq. 3, is still valid [15]. Differentiating Eq. (2) with respect to vector r_{ij} [7], the expression which describes the magnetic force between two magnetic dipoles is obtained:

$$F_{ij}^m(r_{ij}) = -\frac{\partial U_{ij}}{\partial r_{ij}} = -\frac{3}{4\pi\mu_0} \left[\left[5 \frac{(m_i \cdot r_{ij})(m_j \cdot r_{ij})}{|r_{ij}|^7} r_{ij} - \frac{m_i \cdot m_j}{|r_{ij}|^5} r_{ij} - \left\{ \frac{(m_j \cdot r_{ij})m_i + (m_i \cdot r_{ij})m_j}{|r_{ij}|^5} \right\} \right] \right] \quad (5)$$

The behavior of the interacting particles is visualized in Figure 2. If vector r_{ij} , which links the geometrical centers of the particles, is parallel to the vector describing external field intensity H , the particles will attract each other. On the other hand, if vector r_{ij} is perpendicular to vector H , the repulsive force will be present. In other cases, the rotation will take place and then the particles will attract each other.

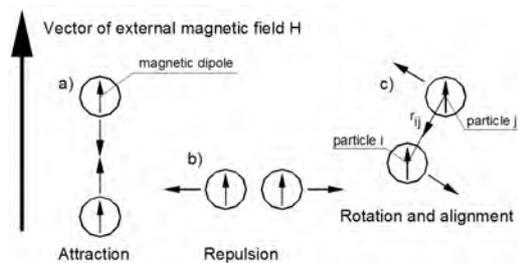


Fig. 2. Behavior of interacting magnetic dipoles [14]

Rys. 2. Oddziaływania magnetostacyjne dipoli [14]

It is worth stressing here that if the particles touch each other and vector r_{ij} is parallel to vector H , the value of the magnetostatic attractive force will be equal to:

$$F_{ij}^{m \text{ ATTR}}(|r_{ij}| = 2a) = 2F_0, \quad F_0 = \frac{3m_i \cdot m_j}{4\pi\mu_0(2a)^4}. \quad (6)$$

However, if the particles touch each other and vector r_{ij} is perpendicular to vector H , the value of the magnetostatic repulsive force will be equal to:

$$F_{ij}^{m \text{ REPUL}}(|r_{ij}| = 2a) = -F_0. \quad (7)$$

It means that the magnetostatic attraction is twice as large as the magnetostatic repulsion.

Additionally, it is possible to obtain from Eq. (2) an expression that describes the torque acting on the magnetic dipoles [7, 20]. Nonetheless, according to the authors' experience, its influence on the final particle configuration is negligible.

Short-range repulsive interactions

As a result of the magnetostatic attraction, the positions of the particles can overlap during the simulations. In order to prevent this, it is necessary to introduce short-range repulsive forces [14]. The ferromagnetic particles are hard spheres, hence the two-body interaction potential of the repulsion should be described in the following way:

$$U_{ij}^{rep} = \begin{cases} \infty, & |r_{ij}| \leq 2a \\ 0, & |r_{ij}| > 2a \end{cases} \quad (8)$$

However, this form of short-range repulsive potential makes MD simulation very difficult or even impossible to carry out. Therefore, a continuous form of the interaction should be introduced. In the presented work, two types of expressions are used in order to define the short repulsive interaction, i.e. polynomial and exponential:

$$\begin{aligned} \text{a) } F_{ij}^{REP} &= -2F_0 \left(\frac{2a}{|r_{ij}|} \right)^n \frac{r_{ij}}{|r_{ij}|}, \\ \text{b) } F_{ij}^{REP} &= -2F_0 \exp \left[-\kappa \left(\frac{|r_{ij}|}{2a} - 1 \right) \right] \end{aligned} \quad (9)$$

where n , κ are the free parameters and F_0 is the constant defined in Eq. (6). A comparison between the short-range repulsive interactions with different values of free parameters n , κ and the magnetostatic interactions (Eq. (5)) is shown in Figure 3.

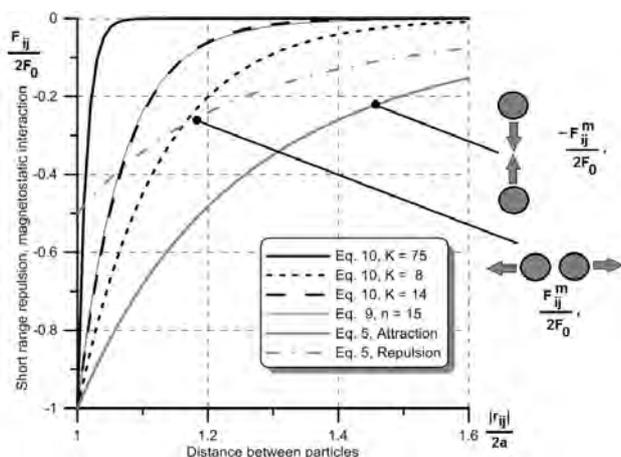


Fig. 3. Short-range repulsion, magnetostatic interactions versus distance between particles

Rys. 3. Oddziaływania krótkozasięgowe oraz magnetostaticzne w funkcji odległości cząstek

It is worth noting that with an increase in the value of parameter κ , the values of the function described in Eq. (9b) more quickly tend towards 0. Moreover, when the values of n and κ are selected suitably, the relationships described by Eq. (9a) and Eq. (9b) are very similar. In that case the results of MD simulations should also be very similar. In contrast, the magnetostatic interactions are quite different.

Hydrodynamic resistance

The molecules of a carrier fluid are incomparably much smaller than the ferromagnetic particles, hence the carrier fluid is considered as a continuous medium. All the interactions between the moving particles and the carrier fluid can be described with the use of the following relationship [14]:

$$F_i^{HYD} = -R_{FU}(v_i - v_\infty) + R_{FE} : E^\infty, \quad (10)$$

where F_i^{HYD} is the generalized force - torque vector, v_i is the generalized transitional - rotational velocity vector, v_∞ is the generalized imposed transitional - rotational velocity vector determined at the center of the ferromagnetic particle, E^∞ is the symmetric part of the velocity gradient tensor. The elements of tensors R_{FU} and R_{FE} depend only on the particle position and describe all the hydrodynamic interactions between the particles. Due to difficulties connected with determining tensors R_{FU} and R_{FE} , very often Eq. (10) is replaced by Stokes' drag:

$$F_i^{HYD} = 6\pi\eta a(v_i - v_\infty), \quad (11)$$

v_i is the transitional velocity vector and v_∞ is the ambient transitional velocity vector determined in the center of the i -th particle. Rotational velocity is typically neglected. η stands for the viscosity of the carried fluid. Nevertheless, this simplification could cause significant errors during the simulation when the hydrodynamic forces begin to dominate over the other interactions. This is very important, especially in the case of a flow simulation. Joung and See [23, 24] used the Rotne-Prager-Yamakawa tensor in order to take a much more complicated form of hydrodynamic resistance force into account. Unfortunately, the proposed method is very time consuming because of the need for matrix inversion, whose size depends on the number of particles. Satoh et al. [20] proposed a numerical procedure to determine the coefficients of the hydrodynamic resistance tensor.

Equation describing particle motion

A change in the acceleration, velocity and position of each ferromagnetic particle is determined by Newton's Second Law:

$$M_i \frac{d^2 r_i}{dt^2} = \left(\sum_{i=1, j \neq i}^N F_{ij}^m + F_{ij}^{rep} \right) + F_i^{hyd}, \quad (12)$$

where M_i denotes the mass of the i -th particle. On the other hand, in the case of MR fluids the inertia force of a moving particle in comparison with the magnetostatic force is several orders of magnitude smaller [15]. Therefore, the left side of Eq. (12) could be ignored. Furthermore, taking Eq. (11) into account, Eq. (12) can be rewritten in the following simplified form:

$$\frac{dr_i}{dt} = v_\infty + \frac{1}{6\pi a_i \eta} \left(\sum_{i=1, j \neq i}^N F_{ij}^m + F_{ij}^{rep} \right), \quad (13)$$

The equation above together with initial condition $r_i(0) = r_0$, is a first order differential equation. Usually, this problem is solved numerically with the use of the very simple Euler's procedure.

RESULTS OF SIMULATION

In the performed simulations the total number of particles is equal to $N = 529$. All the particles are spherical in shape with an identical radius a , where $a = 8e - 6$ m. The length of the square edge of the simulation area is $l_s = 5.55243e - 4$ m. The volume is filled with carrier fluid with viscosity $\eta = 0.1$ Pa s. The value of the volume fraction is evaluated with the use of Eq. (1) and is equal to $v_f = 23\%$. In this case the effective

magnetic permeability is evaluated as $\mu^{eff} = 5.605$. The simulation area is subjected to the action of the uniform external magnetic field with intensity $H_0 = 3e4[A/m]$. The total time of the simulation is assumed as $t_c = 0.5$ s and the time step is constant and $dt = 1e - 6$ s. It means that during the simulation $5e5$ time steps are performed. Moreover, in order to accelerate the simulation procedure, a cut-off radius [7] is introduced and its length is equal to $r_c = 10a$. Four simulations are carried out, namely three simulations where the exponential form of the equation describing the short repulsive interactions (Eq. (9b) with $\kappa = 8, \kappa = 14, \kappa = 75$) is assumed and one simulation where the polynomial form (Eq. (9a) with $n = 15$) is defined. No global flow of the MR fluid is taken into account ($v_\infty \equiv 0$ in Eq. (13)).

Generation of the initial configuration of the ferromagnetic particles consists of two steps. In the first step, the particles are uniformly placed in the simulation area and next the coordinates describing its position are randomly disturbed. Following this procedure, "quasi-uniform" configuration of the particles is obtained.

The obtained results are presented in Figure 4, which illustrates the configurations of the ferromagnetic particles in the case of using Eq. (9b). As can be seen, the process of microstructure formation is very fast, especially in the initial phase.

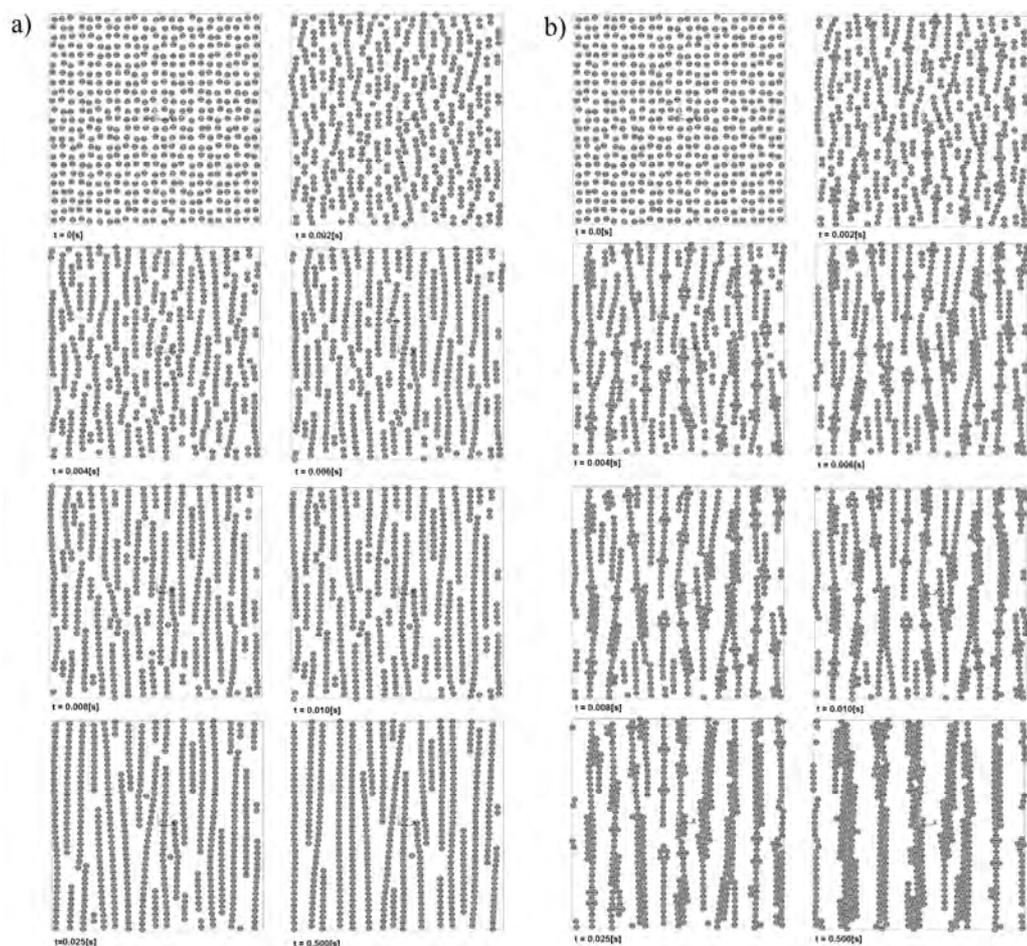


Fig. 4. Results of MD simulation, Eq. (9b), a) $\kappa = 8$, b) $\kappa = 75$

Rys. 4. Wyniki symulacji MD, Eq. (9b), a) $\kappa = 8$, b) $\kappa = 75$

All the internal structures consist of strings of particles which are generally parallel to the direction of the vector of the applied external magnetic field intensity. However, in the case of small κ values, simple isolated chains of ferromagnetic particles are observed (Fig. 4a, $\kappa = 8$). Increasing the κ value leads to a change in the final shape of the microstructure. Now the strings of the particles are more irregular and aggregations of the particles become visible. For large values of parameter κ there are no simple isolated strings of particles. Thick columnar aggregations of particles are then observed (Fig. 4b, $\kappa = 75$).

To summarize the obtained results of the MD simulations, it should be stressed that short-range repulsive interactions have a significant effect on the final shape of the microstructure. Moreover, in the case of a larger parameter κ value, the formation process takes more time. The final configuration obtained with the use of the polynomial form of the short-range repulsive interaction (Eq. (9a), $n = 15$) is very similar to the final shape of microstructure obtained by means of Eq. (9b), $\kappa = 14$, which can be seen in Figure 5.

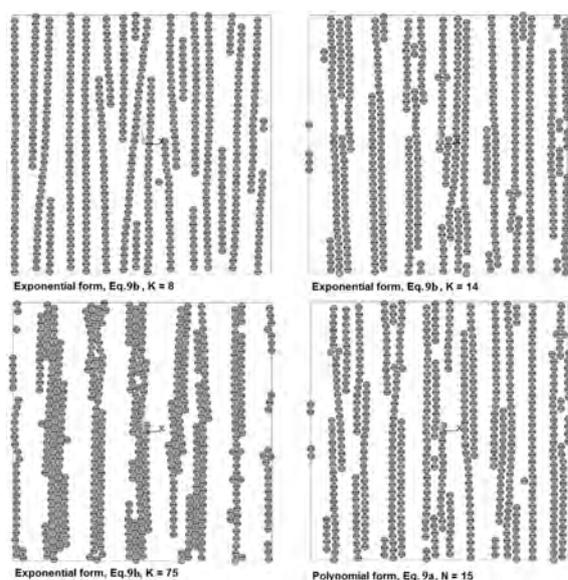


Fig. 5. Comparison of final configuration of ferromagnetic particles

Rys. 5. Porównanie uzyskanych konfiguracji cząstek ferromagnetycznych

CONCLUSIONS

The present paper is devoted to computational simulations of the behavior of ferromagnetic particles under the influence of an external magnetic field in magneto-rheological fluids. The main aim of this work is to investigate the effect of different forms of relationships describing short-range repulsive interactions. Generally, two forms of equations that determine these forces are taken into account, i.e. polynomial and exponential. It was found that the main influence is exerted by the parameter values which determined the effective range of the discussed interactions. For larger values of parameters n (Eq. (9a)) or κ (Eq. (9b)) (shorter effective

range), the final microstructure consists of thick clusters of ferromagnetic particles. This kind of microstructure is observed in the case of real MR fluids. Notwithstanding, for small values of parameters κ or n , the obtained microstructure is quite different; the particles create simple isolated strings.

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