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Introduction

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Heterogeneous materials: metastable and non-ergodic internal structures

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This issue is concerned with structural and phase transitions in heterogeneous and composite materials, the effects of external magnetic fields on these phenomena and the macroscopic properties and behaviour of materials with isotropic and anisotropic internal structures. Using experimental, theoretical and computer methods, these transitions are studied at the atomic and mesoscopic levels. The fundamental specific feature of structural transitions in many heterogeneous media consists of the fact that these transitions are stacked for a long time in non-equilibrium states that appear due to either macroscopic dissipative processes (an alternating magnetic field or hydrodynamic flow, for instance) or system lifetime in a metastable state. It is important to explain and describe these transitional states using the general approach of non-equilibrium physical mechanics. The review and research articles in the issue will cover the whole spectrum of scales (from nano to macro) and materials (from metastable liquids to biological polymers) in order to exhibit recently developed trends in the field of heterogeneous materials. Atomistic modelling, structuring induced by external magnetic fields and hydrodynamic flows, metastable and non-ergodic states, mechanical properties and phenomena in heterogeneous materials—all these are covered.

This article is part of the theme issue 'Heterogeneous materials: metastable and non-ergodic internal structures'.

1. Introduction

Heterogeneous materials are widespread in Nature (soils, geological formations, biological tissues) and actively

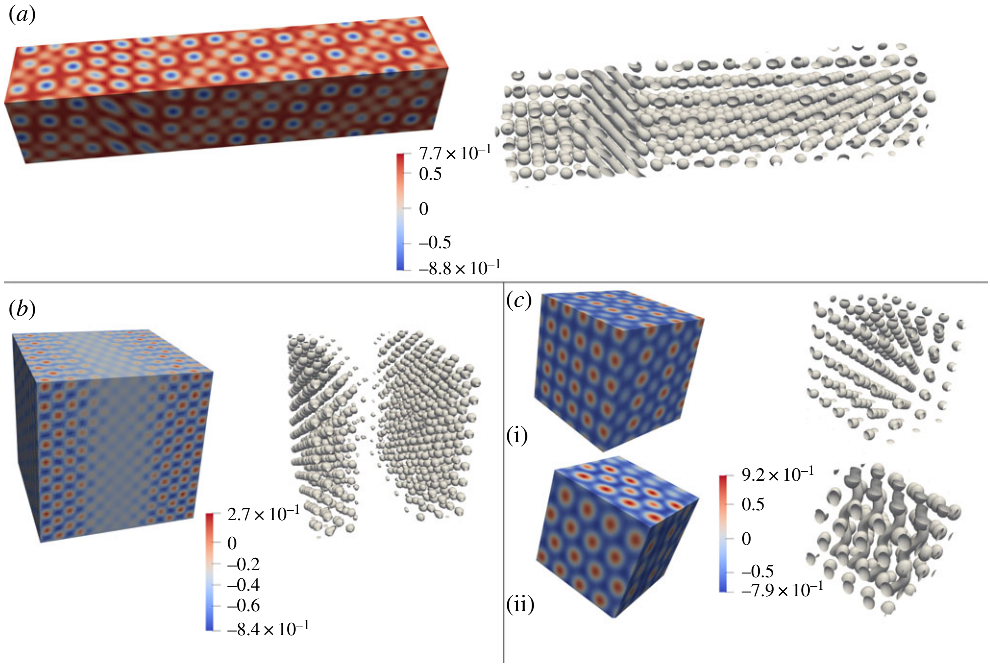

Figure 1. Snapshots of crystalline structures simulated by solution of hyperbolic phase-field crystal model (the left panels represent the distributions of the atomic density on the surfaces of computational domains; the right panels show the isosurfaces of the average atomic density in a volume of computational domain): (*a*) the crystalline structure with coexistence of two types of patterns (body-centred cubic and rods) for $\varepsilon = 0.5$; (*b*) the evolution of the face-centred cubic pattern structure for $\varepsilon = 0.5$; (*c*) (i) the stable body-centred cubic structure for $\varepsilon = 0.4$, and (ii) the irregular metastable formation for $\varepsilon = 0.4$ (ε is the dimensionless undercooling). The colour bars mean the legends of the atomic density. The calculations were carried out by Ilya Starodumov and are presented in more detail in [1].

used in many modern industrial and biomedical technologies as advanced smart and composite materials. The macroscopic properties and behaviour of these systems are determined by internal structures formed by nano- and micrometre-sized inclusions in the host material or as a result of various phase transitions in these systems (see e.g. figure 1). In part, magnetic soft and liquid composites (magnetic gels and elastomers, magnetic suspensions) attract especial interest because of the possibility to control their physical and mechanical properties and macroscopic behaviour by using a magnetic field, which is quite achievable in laboratories and practical conditions. This is why the analysis of the evolutionary kinetics of these structures and their stable and metastable states requires more detailed theoretical and experimental study. Experiments show that very often internal structures in materials do not achieve their thermodynamic equilibrium state at the time of observation and are stacked in 'frozen' metastable or other transition states. A hydrodynamic flow in dense suspensions and other complex fluids can provoke the appearance of specific internal heterogeneous structures that significantly affect the rheological and other properties of these fluids. For example, they can increase viscosity by several orders of magnitude. This is why the analysis of the kinetics of the evolution of these structures and their stable and metastable states requires detailed theoretical and experimental study.

In this issue, we demonstrate the whole physical spectrum of the formation and evolution of internal structures in non-organic and biological heterogeneous materials, from the atomic level of newly born crystals to their growth at the mesoscopic level, structural transformations in ensembles of nano- and micrometre-sized particles and, finally, the effect of these structures on the macroscopic physical and rheological properties of heterogeneous systems. The present works include experimental, theoretical and computer studies of the aforementioned systems and phenomena. This issue builds upon previous works regarding structure formation developed by a number of famous scientists. New research articles devoted to cutting-edge results in particular areas of expertise form the basis of this issue. The issue is structured in accordance with article themes, moving from atoms to mesoscopic metastable, non-ergodic and dissipative structures and vitally important biomedical applications. This feature reflects the tendency to describe internal structures in various heterogeneous materials at different scales and represents an original approach on the part of this issue. In general, the present theme issue covers new developments in the structuring of materials, through metastable, non-ergodic and non-equilibrium transitions to different internal formations in heterogeneous materials of broad applied interest (ranging from condensed matter physics and the chemical industry to biomedical technologies and the life sciences).

The present issue covers the rapidly developing research area of heterogeneous materials obtained through metastable and non-ergodic structural and phase states. It covers multiple disciplines, ranging from condensed matter physics and materials science to biomedical applications. To present a complex picture of micro- (nano-) and meso-temporal and spatial scales in structural and phase transitions, a study of various heterogeneous materials (from metastable liquids to composites of biopolymers) is included in this issue. Special attention is paid to developments in the scientific background of these materials' biomedical applications (the polymerization and crystallization of proteins and haemoglobins, magneto-controlled biogel scaffolds for the regeneration and engineering of biological tissues, the method of magnetic hyperthermia used in the therapy of oncological diseases). On the atomistic scale, computational and theoretical models of the kinetics of liquid–solid phase transitions are presented. On the mesoscale, advanced models of the evolution of particulate ensembles in metastable liquids are considered. Experimental and theoretical studies of dissipative, hydrodynamically provoked structurization in suspensions of magnetic particles are included.

2. The general content of the issue

Heterogeneous systems and smart materials are widespread in Nature and actively used in many industrial and biomedical technologies. The internal phase and structural transformations in these systems (for instance, crystal growth in liquids, the aggregation of fine particles of fillers in composite materials, the crystallization of haemoglobin C and the polymerization of haemoglobin S, the crystallization of proteins in the retina responsible for the formation of cataracts, interferon-alpha and human growth hormone synthesis, etc.) affect and determine their macroscopic properties and behaviour. Such structures arise during different phase and structural transformation processes, ranging from materials physics to the life sciences. By using different external and combined processes (magnetic and electric fields, hydrodynamic flows, crystallization combined with polymerization, which, in the case of haemoglobins C and S, reduces the plasticity of the body's erythrocytes and causes sickle cell anaemia, and so on), one can tune the scenario of these transformations and the final morphology of internal structures. In its turn, this opens up the possibility of controlling the macroscopic properties and behaviour of these materials with the help of external fields and other impacts.

Recently, it has been shown that structures in non-equilibrium and non-ergodic transitions not only evolve according to the classical theory of phase transitions but also can be provoked by external dissipative impacts (hydrodynamic flows or the process of polymerization, for instance) [2], leading to different unusual states: consider, for example, long-lived glass-like structures [3,4] or protein crystallization [5–7]. Study of these dissipative and non-ergodic structures has opened up in modern physics the new and challenging field of condensed matter and smart materials. In this issue, the multi-scale problems of metastable, dissipative and non-ergodic structures in heterogeneous systems and materials, as well as some problems surrounding their biomedical applications, are considered. The issue is compiled using the naturally arising hierarchic principle, moving from the atomic level via mesoscopic description to the macroscopic properties of materials.

(a) The atomic-scale level

The atomic-scale level of description is based on computer study and modelling of the kinetics of phase transition and the structural evolution at the phase transition interface. Several numerical models and methods for their realization in the evolution of heterogeneous structures in solidified materials are considered in this part of the issue. An overview of modern computational methods designed for phase-transition problems is presented in the review paper of Hector Gomez, Miguel Bures & Adrian Moure [8]. This review will help the community to understand the new computational challenges brought by phase-transition problems and will provide tools to efficiently study phase transitions in physics. Here, such modern computational methods as the phase-field crystal method and the isogeometric analysis required for the spatial discretization of the modified phase-field crystal model are discussed and applied. A new approach continuing this review paper on thermodynamics of rapid solidification and crystal growth kinetics in glass-forming alloys is developed in the next paper given by Peter Galenko *et al.* [9], where a deviation from thermodynamic equilibrium is introduced via gradient flow relaxation of the phase field. Another type of heterogeneity in the form of solute channels (so-called freckles) is considered by Andrew Kao *et al.* [10]. These freckles are common defects which appear during the solidification of metal alloys. A numerical model of these defects and methods for the realization of the model are considered in this paper. Moving on from the atomic level of non-equilibrium and non-ergodic phase transitions, we gradually go to the mesoscopic and macroscopic levels, where the results of previous papers are used when considering the evolution of particulate ensembles in metastable liquids, magnetic suspensions and gels.

(b) Metastable and non-ergodic systems governed by macroscopic (external) impacts

Here, we consider phase transition processes and phenomena occurring in non-equilibrium systems such as deeply undercooled liquids and supersaturated solutions under the influence of different additional factors representing driving forces and leading to unusual dynamical behaviour. A theoretical study of liquid phase separation in undercooled alloys under the conditions of static magnetic fields is presented by Dandan Zhao & Jianrong Gao [11]. Here it was found that the imposition of static magnetic fields can alter the morphology, segregation and size distribution of droplets due to liquid phase separation. Undercooling experiments, analytical calculations and phase-field simulations for Ni–Fe–Si alloy systems are developed by Dasari Mohan & Gandham Phanikumar [12], along with new insights concerning the presence of additional impurities (undercooled ternary systems). The parametric space for dendrite growth is explored to understand the fundamental features of phase transition. The results are discussed in light of current non-equilibrium models. Experiments on the microstructural evolution of eutectic alloys processed at different cooling rates are carried out by Olga Gusakova *et al.* [13]. The authors found that a coupled eutectic growth leads to the formation of lamellar structures. The paper presented by Dmitri Alexandrov & Irina Alexandrova [14] is devoted to the critical question of what mechanisms control particle growth rates in their initial and intermediate growth stages in metastable liquids. In this paper, a new theory of the unsteady-state thermo-solutal growth of spherical particles in a metastable surrounding is developed so that the main non-stationary corrections produced by fluctuations in the fundamental contribution of the particle's growth rate are found. This paper, which is directly connected with experiments and simulations presented in papers [11,12], opens a new perspective on the theory of phase transformations at the intermediate stage, which is presented in the next paper by Eugenya Makoveeva & Dmitri Alexandrov [15]. In this paper, a new theory describing the evolution of a phase transformation process in supersaturated and supercooled liquids is considered with allowance for the fluctuation corrections in particle's growth rates [14]. Note that some important applications of this theory in the life sciences and biophysics are considered in the last subsection of this issue.

Liquid and soft composite materials with nano- and micrometre-sized particles undergo dissipative and non-ergodic phase transitions that determine the macroscopic properties and behaviour of these systems. Percolating net-like structures provoked by hydrodynamic forces in macroscopically flowing concentrated magnetic suspensions are considered experimentally and theoretically in the paper presented by Georges Bossis *et al.* [16]. The results of this work demonstrate that the appearance of these structures leads to a discontinuous jump upwards of suspension viscosity and to auto-oscillations in flow when the macroscopic shear rate achieves some threshold magnitudes. These effects can be either useful or harmful from the point of view of various technologies based on dense suspensions (building, pharmaceutical, perfume, food industries, etc.). The usage of magnetic particles in these systems opens the prospect of controlling rheological phenomena in these systems with the help of an applied magnetic field.

Magnetopolymer composites, consisting of a gel or elastomer matrix and embedded magnetic nano- or micrometre size particles, present a new kind of smart materials, which are actively used in various progressive industrial and biomedical applications. Macroscopic physical properties of these materials are determined by the morphology of internal structures, formed by the particles of the filler. In part, one can control, in a wide range, the mechanical characteristics and behaviour of the composites by using an external magnetic field. Anisotropic structures in the magnetic polymers with a high concentration of magnetic particles, the effect of these structures, as well as the effect of external magnetic field on the mechanical properties of the composites are studied in the work reported by Dmitry Borin, Gennady Stepanov and Eike Dohmen [17]. The last two papers shed light on the internal structures in highly concentrated magnetic composites with liquid and mechanically soft matrices as well as on macroscopical properties of these systems. Experiments (see, for example, papers [3,4] as well as paper [17] in this issue) show that non-ergodic long-lived structures appear in magnetic gels and suspensions under the action of an applied magnetic field. These structures determine the macroscopic properties and behaviour of these composite systems. In part, they lead to variation, in the range of several orders of magnitudes, in rheological, electronic and other characteristics of the composites. In turn, this offers the opportunity to control the behaviour of these systems by using an external magnetic field.

(c) Biomedical applications of heterogeneous materials

This subsection is devoted to some important practical applications of the approaches developed in the two previous subsections. In this subsection, we discuss the rapidly developing biomedical research field of heterogeneous materials obtained through metastable and non-ergodic transitions and states. Such vital applications as the synthesis of proteins, the crystallization and polymerization of haemoglobins, magnetic hyperthermia and biocompatible hydrogels represent the main subjects of the present theme issue. Obtaining new results in various biochemical and biomedical research is hindered by the lack of a complete understanding of the mechanisms of the nucleation and growth of proteins. For example, a slow dissolution rate of protein crystals is used to produce drugs such as insulin. Note that the synthesis of other proteins (e.g., interferon-alpha and human growth hormones) is also based on the nucleation process. Here, an important role is played by the following circumstance. If the administered dose consists of several large crystals, the release rate of the drug can be maintained for a longer time than if the dose consisted of a large number of smaller crystals. To achieve such size distributions, the nucleation time of the crystals must be small so that all the crystals grow with almost identical supersaturation. It is possible to mention other biomedical applications that arise, for example, when pathological conditions are associated with the formation of crystals or other solid aggregates in the human body. An important example is the crystallization of haemoglobin C and the polymerization of haemoglobin S, which reduce the plasticity of the body's erythrocytes and cause sickle cell anaemia. Another important example is the crystallization of proteins in the retina, which is responsible for the formation of cataracts. Taking into account the high importance of describing phase transformations in proteins, the developed theory is compared with experimental data on protein crystallization. Namely, the paper presented by Dmitri Alexandrov & Irina Nizovtseva [18] is devoted to the theoretical development of the nucleation processes and connects the theory

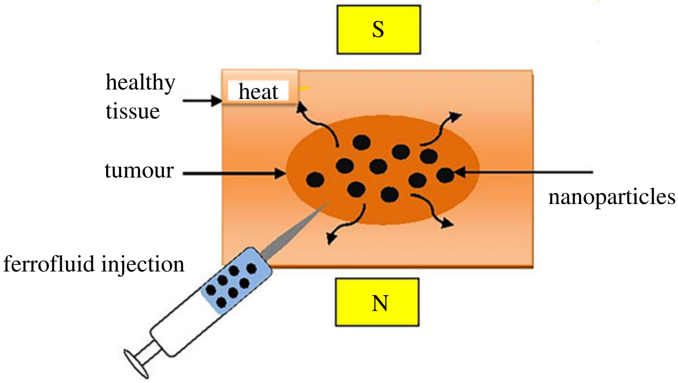

Figure 2. Illustration of the magnetic hyperthermia application. Magnetic nanoparticles are injected into the tumour region and heated there by alternating magnetic field. (Online version in colour.)

and experimental data on protein and insulin crystallization. Specifically, this paper develops the previous papers [14,15] to describe the effect of the 'diffusion' of the particle-size distribution function in the space of particle radii. In addition, the obtained and generalized dynamical laws are used here to describe and explain existing experiments for biomedical applications. The next paper, presented by Alexandr Ivanov, Irina Alexandrova & Dmitri Alexandrov [19], develops the theory of previous papers to include the effect of combined nucleation, growth and polymerization at the particulate level. Here, a new theory leaning on modified integro-differential kinetic and balance equations is formulated. New solutions founded on the basis of the saddle-point method for the Laplace-type integral are constructed to describe a new class of phase transformation processes and phenomena occurring in the presence of polymerization. Note that the practical importance of developing these new theoretical approaches of non-equilibrium phase transformations is considerable for the field of materials physics, production of specialty chemicals and biomedical applications.

Magnetic hyperthermia (the heating of a medium by embedded magnetic nanoparticles under the action of an alternating magnetic field) is a prospective method of therapy for oncological diseases [20–22] (see also figure 2). The development of clinical applications for this method requires a detailed study of the mechanisms of heat production in the systems of the injected particles. The absolute majority of works in this field deal with model systems of single, non-interacting particles. However, experiments show that the particles can be accumulated in certain places in biological cells. This is why analysis of their cooperative reaction to an applied magnetic field, as well as the development of methods of mathematical prediction for heat production, is an important and relevant problem. The effect of magnetic interparticle interaction on the neat production in the systems of immobilized particles, united in chain-like structures and uniformly distributed in a host medium, is studied by Andrey Zubarev in [23] and by Ali Fathi Abu-Bakr & Andrey Zubarev in [24], respectively.

The damage, failure or loss of a tissue or organ is a common problem that affects most human beings at least once in their lifetime. When the body is unable to heal itself, traditional approaches to this issue include autologous or allogeneic transplantation. Unfortunately, the availability of autologous tissue is limited and there is a non-negligible risk of immunorejection of allogeneic sources. As a result, the mismatch between the number of patients waiting for transplantation and the supply of organs and tissues is continually growing, which constitutes an unresolved health issue of increasing magnitude across the world. Tissue engineering is an interdisciplinary field that applies the principles of biology and engineering for the development of strategies aimed at the replacement, repair, maintenance and/or enhancement of tissue function. Biocompatible hydrogels are actively used in bioengineering and clinical medicine as scaffolds for manufacturing, curing and regenerating biological tissues [25,26] (see also figure 3).

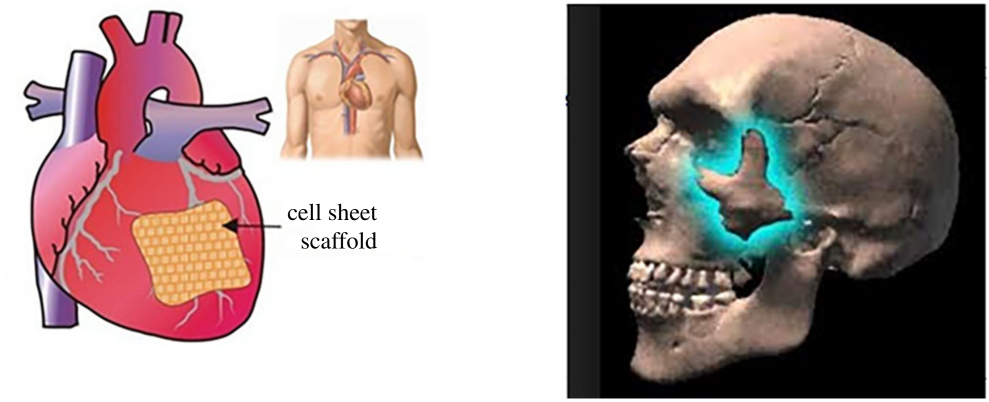

Figure 3. Examples of the medical application of the biocompatible hydrogel scaffolds in cardiology and for skull regeneration.

Scaffolds made of magnetic hydrogels have several significant advantages compared to their non-magnetic analogues. The internal structures and rheological properties of biological hydrogels with embedded spherical and rod-shaped magnetic particles are studied by Cristina Gila-Vilchez *et al.* [27] and by Mariem Abrougui *et al.* [28]. The aim of these works is to develop the scientific background for the preparation and usage of magnetic scaffolds in engineering and the regeneration of biological tissues.

3. Conclusion

Recently, a large number of highly important works have been completed in the experimental study and atomic and mesoscopic modelling of structural and phase transitions in heterogeneous media and materials. Among them are works devoted to the influence of magnetic fields and hydrodynamic flows on the internal structures in these systems, the combined influence of nucleation and polymerization on the evolution of metastable materials, phase and structural transitions and the effect of internal structures on the macroscopic properties and behaviour of heterogeneous systems (rheological phenomena, the features of biological tissue growth and cell proliferation, the formation of a monocrystal or polycrystal internal structure). This issue is focused on new experimental and theoretical advances in the field of metastable, non-ergodic and dissipative structures in heterogeneous media and composite materials. This issue presents overviews and regular articles on experimental, computational and theoretical studies of non-equilibrum and non-ergodic phase and structural transformations in heterogeneous materials. We hope that this issue is important for a wide audience, including scientists, engineers, lecturers, postgraduates and undergraduates, due to the educational and practical value of the included articles. An important point of our theme issue is the wider public interest in some of the vital biomedical applications of structural and phase transformations in metastable and non-ergodic heterogeneous materials. Applications like the synthesis of proteins, the polymerization and crystallization of haemoglobins, magnetic hyperthermia and biocompatible hydrogels may be mentioned, among others.

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Competing interests. The authors declare that they have no competing interests.
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