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An Arlequin-based method to couple molecular dynamics and finite element simulations of amorphous polymers and nanocomposites

S. Pfaller [a], M. Rahimi [b,1], G. Possart [a], P. Steinmann [a,*], F. Müller-Plathe [b,1], M.C. Böhm [b,1]

[a] Chair of Applied Mechanics, University of Erlangen-Nuremberg, Egerlandstr. 5, 91058 Erlangen, Germany
[b] Eduard-Zintl-Institut für Anorganische und Physikalische Chemie and Centre of Smart Interfaces, Technische Universität Darmstadt, Petersenstr. 20, 64287 Darmstadt, Germany

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ABSTRACT

A new simulation technique is introduced to couple a flexible particle domain as encountered in soft-matter systems and a continuum which is solved by the Finite Element (FE) method. The particle domain is simulated by a molecular dynamics (MD) method in coarse grained (CG) representation. On the basis of computational experiences from a previous study, a staggered coupling procedure has been chosen. The proposed MD–FE coupling approximates the continuum as a static region while the MD particle space is treated as a dynamical ensemble. The information transfer between MD and FE domains is realized by a coupling region which contains, in particular, additional auxiliary particles, so-called anchor points. Each anchor point is harmonically bonded to a standard MD particle in the coupling region. This type of interaction offers a straightforward access to force gradients at the anchor points that are required in the developed hybrid approach. Time-averaged forces and force gradients from the MD domain are transmitted to the continuum. A static coupling procedure, based on the Arlequin framework, between the FE domain and the anchor points provides new anchor point positions in the MD–FE coupling region. The capability of the new simulation procedure has been quantified for an atactic polystyrene (PS) sample and for a PS-silica nanocomposite, both simulated in CG representation. Numerical data are given in the linear elastic regime which is conserved up to 3% strain. The convergence of the MD–FE coupling procedure has been demonstrated for quantities such as reaction forces or the Cauchy stress which have been determined both in the bare FE domain and in the coupled system. Possible applications of the hybrid method are shortly mentioned.

1. Introduction and outline

Continuum mechanics, a field-based theory that can be treated numerically by the widely used Finite Element (FE) method, is a very powerful tool to simulate the macroscopic mechanical behavior of e.g. solids. Unfortunately, it is less suited in cases where molecular or even atomistic details become important to capture the phenomena of interest. On the other hand, such particle-based information becomes accessible either by Monte Carlo (MC) or molecular dynamics (MD) calculations. The application of MC or MD simulations for macroscale problems, however, is computationally prohibitive due to the large number of particles required and the corresponding degrees of freedom. Typical system sizes in MD calculations are in the range of nanometers, while typical orders of magnitude in the time steps amount to femtoseconds, which allow simulation times of some nanoseconds. Compared to the time scales and system sizes relevant in engineering problems of continuum mechanics, the time and length scales considered in MD simulations are, thus, orders of magnitude smaller. The advantages of both theoretical tools can be combined within hybrid techniques which allow the coupling of a particle description to a continuum one or the coupling of particle domains with different resolutions. Such methods are particularly useful under conditions where molecular details are relevant only in smaller spatial regions of the studied sample such as solid–fluid or solid–polymer interfaces, while particle properties are not required in the remaining regions which have bulk behavior. In such systems the efficiency of continuum mechanics can be combined with the accuracy of MD simulations.

In the past years an increasing number of publications have proposed combinations of particle-based models with field-based models in hybrid schemes. Within this contribution, we will concentrate on hybrid schemes which are based on a spatial decomposition into a particle region and a much larger continuum. Unlike

* Corresponding author.
E-mail addresses: sebastian.pfaller@ltm.uni-erlangen.de (S. Pfaller), m.rahimi@theo.chemie.tu-darmstadt.de (M. Rahimi), gunnar.possart@ltm.uni-erlangen.de (G. Possart), paul.steinmann@ltm.uni-erlangen.de (P. Steinmann), f.mueller-plathe@theo.chemie.tu-darmstadt.de (F. Müller-Plathe), boehm@theo.chemie.tu-darmstadt.de (M.C. Böhm).
[1] Tel.: +49 6151 16 6523; fax: +49 6151 16 6526.

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this, so-called parameter inheritance schemes have become more popular since they can be formulated more easily. These methods allow computing material parameters at the molecular level and use them as input in the subsequent continuum simulations. Quite similarly, single-chain-in-mean-field models have been developed, cf. e.g. [1–3]. Hybrid approaches can be divided into a group for hydrodynamics problems and one for structural mechanics problems. We will focus here on the second type, which often deal with the failure of crystalline systems. However, there are only few adaptions to amorphous systems, which we will concentrate on in this contribution. Well-known examples are the quasi-continuum methods as they were introduced and employed e.g. in [4–6]. Here, selected particles are moved by an affine deformation of the elements since they are attached to FE nodes or edges.

Beyond these kinds of coupling methods, extensions towards amorphous materials have been made by the so-called bridging domain method introduced by Belytschko and co-workers e.g. in [7,8], employed in modeling studies of carbon nanotubes as well as other systems, cf. [9,10]. Quite similar, Ben Dhia and Rateau developed a hybrid scheme called the Arlequin method, cf. [11–13]. Originally used as a tool to couple FE domains of different resolutions, the Arlequin method has been employed later in hybrid simulations of atomistic and continuum models, cf. e.g. [14].

The Arlequin method considers a pure particle region without any underlying FE description, i.e. it does not require the particles to be arranged in a lattice. Instead, a handshake region is introduced in which the FE domain and the particle region overlap. Here the deformation of the continuum and the displacements of the particles are required to match in a weak sense. Thus, it is a suitable candidate to model amorphous materials like polymers. Although there is already a small number of publications available dealing with generic polymer models, by e.g. the group of Prudhomme, cf. [15,16], no procedure has been described yet that allows a coupling between an FE domain and a particle system computed with the commonly used MD tools at finite temperature.

As mentioned above, engineering problems captured by continuum mechanics and microscopic considerations treated by particle-based methods differ tremendously in time and length scales. Thus, both aspects have to be considered carefully here: on the one hand, the spatial scale bridging as introduced here seems to be feasible. Its formulation is supported by the publications on static continuum-particle coupling procedures discussed above. On the other hand, the coupling of time scales that are orders of magnitude different is a very crucial part of any coupling scheme, and it has not been solved yet. In this context, we refer to the group of de Borst and co-workers, which has investigated the spatial and temporal coupling and discussed its advantages and drawbacks in [17].

In this contribution, we have developed a new hybrid technique for polymers to couple a particle domain to a continuum. In order to avoid conceptual problems arising from the different time scales in the respective regions, the continuum is treated purely static while only the particle domain is computed dynamically. This seems to be reasonable due to the large difference between the dynamics on the macroscale and that of the particles: any time-dependent processes on a scale relevant for engineering problems would be quasi-static compared to the dynamics at the level of particles. In the following, we will sketch the main components of the coupling procedure and point to the corresponding sections where these aspects will be discussed in detail.

First, the MD domain treated in the present coupling scheme is large in comparison to dimensions usually encountered in atomistic MD simulations. Therefore the MD region has to be treated by a coarse grained (CG) technique [18–20] which reduces the degrees of freedom by grouping a number of atoms together into so-called superatoms or CG beads. The CG potentials used in the present work have been derived by iterative Boltzmann inversion (IBI) of atomistic potentials [21]. Quite generally the CG mapping offers access to simulations which are computationally unfeasible in a purely atomistic model. To perform coupled MD–FE simulations the conventional periodic boundary conditions (PBCs) had to be replaced by nonperiodic stochastic ones (SBCs) [22]. In the boundary region of the developed coupling scheme we have defined a set of auxiliary particles, so-called anchor points. They are harmonically coupled to the MD particles. The anchor points form a set of fixed particles without interaction between them that do not move during the MD equilibration procedure. Hence, they can be coupled to a static continuum as mediators between the different domains. This static coupling will be realized with the help of the Arlequin method mentioned above.

Thus, the spatial set-up of our hybrid method consists of three main regions as sketched in Fig. 1:

1. a pure particle domain Ω^d;
2. a bridging domain Ω^b where the particle region overlaps with the continuum. This region contains the anchor points;
3. a pure continuum Ω^c, discretized by finite elements.

The scale bridging procedure proposed here is subdivided into a static coupling between the anchor points and the continuum (cf. part (b) in Fig. 1) and into its counterpart which tethers the anchor points to the particle domain (cf. part (c) in Fig. 1). We will subsequently discuss the respective schemes as follows:

- Sections 2.1 and 2.2: dynamic coupling between anchor points and particles as well as necessary modifications of the particle domain
- Sections 2.2, 2.3.1, and 2.3.1: static coupling between anchor points and the continuum, introduction of the Arlequin method

It has to be remarked here that, due to the subdivision into a static and a dynamic scheme, the usual tools to solve the continuum and the particle domain, i.e. the conventional FE and MD procedures, can be applied with rather small modifications. Thus, algorithms and machines highly specialized for the respective tools can be employed which is important especially with respect to the very time consuming MD simulations.

In order to embed both coupling schemes into a single computational procedure, a staggered algorithm as described below has been developed. Within a previous contribution [23], a systematic overview of staggered solution schemes has been given for the hypothetical case of a one-dimensional coupling between a continuum solved with FE and a particle domain mapped by MD. This was aiming at a sound understanding and possible solution of the problems which may appear in such couplings. The results of this examination are used and extended in the present work to couple more realistic systems, i.e. three-dimensional polymer samples as well as nanocomposites containing a polymer component. To render this possible, we had to introduce auxiliary particles, so-called anchor points, which realize a transfer of information between continuum and particle domain. This ansatz for three-dimensional amorphous polymer and composite samples is by no means trivial, at least if compared to crystalline solids, for which hybrid schemes are easier to implement since the particles are spatially confined to a lattice. For the description of the continuum, the theoretical derivations are quite similar to those in [23], whereas the treatment of the particle domain had to be modified significantly. In order to provide a comprehensive description of the methodology, all relevant aspects are recalled subsequently.

Without going into detail, only the conceptual ideas of the staggered scheme are discussed here, for a detailed description we refer to Section 3 and Fig. 5. After applying a load to the undeformed continuum, the resulting deformation is transferred via the