XVI International Conference on Computational Plasticity. Fundamentals and Applications
COMPLAS 2021
E. Oñate, D. Peric, M. Chiumenti and Eduardo de Souza Neto (Eds)

A CONCURRENT MD-FE COUPLING METHOD TOWARDS SIMULATIONS OF FRACTURE OF THERMOPLASTIC POLYMERS

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Key words: Multiscale simulations, MD-FE coupling, Polymer fracture, Thermoplastic polymers, viscoplasticity

Abstract. In this contribution we present a multiscale partitioned-domain method to simulate fracture of amorphous thermoplastic polymers. This method couples a particle-domain treated by the molecular dynamics (MD) framework and a continuum domain discretized by the Finite Element (FE) method. The FE domain is described by a viscoelastic-viscoplastic constitutive model that can reproduce the rate-dependent mechanical behavior of the MD domain within a moderate deformation range. In the highly deformed MD domain near the crack tip, hybrid boundary conditions are applied to mimic the plane strain condition, where the surfaces in the X-Y plane are subjected to periodic boundary conditions while non-periodic stochastic boundary conditions are used for the other surfaces. Using this method, we implement multiscale simulations of polymers with different pre-cracks and different strain rates under Mode-I loading conditions to demonstrate its capability for the investigation of polymer fracture.

1 Introduction

Fracture phenomena typically comprise different scales with specific atomistic and molecular processes near the crack tip requiring an appropriate fine-scale treatment. In contrast, macroscopic boundary conditions form the framework for fracture simulations and are usually based on a continuum mechanical description. Thus, particle-continuum coupling multiscale simulations that resolve the vicinity of the crack tip at particle resolution while describe the remaining part using continuum mechanics are well-suited to investigate fracture of materials.

Although there have been various such methods proposed in the past decades, most of them are not designed for polymers. The pioneering work is the quasicontinuum method [18], followed by the bridging scale method [19], the bridging domain method [3, 20], and the Arlequin method [1] to name a few, together with their applications in simulations of fracture [5, 6, 7, 4].

Since polymers have amorphous structures and a complex topology due to their long chains with entanglements or cross-links, special treatments of the particle domain and the associated coupling strategies are required. To this end, the Capriccio method [15, 13] introduces non-periodic stochastic boundary conditions (SBC) for the molecular dynamics (MD) simulation box, in order to embed it into a continuum domain enabling arbitrary deformations. In addition, auxiliary artificial particles, denoted as anchor points, are inserted into the boundary of the MD domain to apply, among others, an appropriate thermodynamical boundary conditions and to exchange forces and displacements between the domains involved. With these crucial adaptations, the Arlequin method [1] has been extended for simulations of amorphous polymers [13, 11]. The Capriccio method has been used to study the interphase effects of nanocomposites [12, 10, 17] while its framework for simulations of fracture of thermosets in [9].

In its initial version, the Capriccio method uses an elastic constitutive model in the continuum domain, such that it can only capture the mechanical behavior of polymers within a rather small deformation range. Under larger deformation, polymers usually exhibit rate-dependent behavior, which requires a more sophisticated constitutive model. To this end, the Capriccio method has been extended to inelasticity in [23] by employing a viscoelastic-viscoplastic (VE-VP) constitutive model [22], which has been developed besed on MD simulations of coarse-grained polystyrene (PS) in [16]. Since the VE-VP constitutive model is rate-dependent, a temporal coupling between the MD and the continuum domain is introduced to enable simulations with different strain rates.

In the present contribution, the Capriccio method [23] is modified to simulate fracture of thermoplastic polymers under Mode-I deformation condition. A pre-crack is defined in the coupled system by modifying the elements in the continuum domain and deleting corresponding bonds or atoms in the MD domain. Furthermore, hybrid boundary conditions are used in the MD domain where PBC are applied to the surfaces in the X-Y plane while SBC are used for the other surfaces. Using this method, which is described in detail in Section 2, we compare the mechanical behavior of a PS sample with different pre-cracks in Section 3. The current limitations of this method are discussed together with an outlook in Section 4.

2 Methods

2.1 The Capriccio method

The Capriccio method, representing "**C**oupled **A**rlequin-based **P**ar**ti**cle-**C**ontinuum **C**omputat**io**n", couples an MD domain and a continuum domain solved by the Finite Element (FE) method, which overlap in the bridging domain as depicted in Figure 1.

The MD domain is embedded into the FE domain by switching the PBC to SBC [15], where random dissipative forces are added to the outer layer of the MD domain to provide a heat bath and reduce the wave propagation. In addition, anchor points are introduced into the boundary layer which are tethered harmonically to specific super atoms and held fixed during MD simulations to provide thermodynamical boundary conditions to the MD domain.

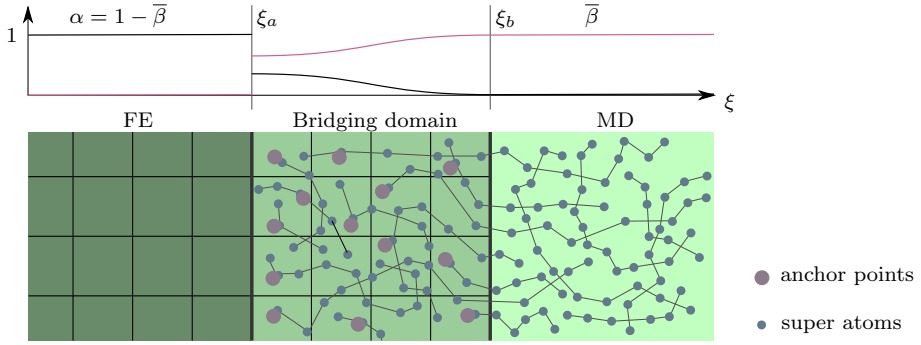

Figure 1: Spatial coupling of the Capriccio method [11, 23]: The FE and MD domain overlap in the bridging domain. The energies are mixed with the weighting factor α while the kinetic and kinematic information is transferred by the anchor points.

The MD domain with SBC is coupled with the FE domain based on the framework of the Arlequin method [1], where the energy of the FE and MD domain are blended with the weighting factor α. However, it is not possible yet to weight our MD system for polymers with pair, bonded, and angular potentials such that a real weighting factor cannot be introduced in the MD domain. This problem has been bypassed in [11] by considering the reduced stiffness due to chain cutting as a weighting effect, where an imaginary weighting factor $\overline{\beta}$ for the MD domain is assumed as illustrated in Figure 1. Consequently, the weighting factor of the FE domain can be calculated as $\alpha = 1 - \overline{\beta}$.

The FE and MD domain transfer information through the anchor points [13], which move according to the deformation of the FE domain based on a Moving Least Square approximation [2]. Since the superatoms in the MD domain are not directly coupled to the FE domain, different domains can be calculated separately with a staggered scheme [13], which has been simplified from 3 stages to 2 stages in [23]: In each load step with a predefined incremental strain $\varepsilon^{\mathrm{ls}}$ of the Dirichlet boundary of the FE domain, the MD domain runs n^{MD} steps. During the calculation of the MD domain, the anchor points are kept fixed and the averaged forces exerted on the anchor points are passed to the FE domain for its subsequent calculation.

To capture the inelastic behavior of the polymer at larger deformation, a VE-VP constitutive model [21] is employed in [23]. Its rate-dependence introduces time derivatives into the FE domain and enables a temporal coupling between the FE and MD domain. Thus, coupled simulations with different strain rates become possible.

2.2 Modification for fracture simulations

In the present contribution, we modify the Capriccio method for multiscale simulations of fracture under Mode-I deformation conditions. We consider coupled systems with precracks as depicted in Figure 2 (a), where an MD domain shown in Figure 2 (b) is enclosed by an FE domain. The system is simulated by prescribing the displacements $u(t)$ of the surfaces in the Y-Z plane at time t. Analogous to plane strain conditions, hybrid boundary conditions are introduced into the MD domain, where the SBC are used for the surfaces

in the X-Z and Y-Z planes while the PBC are employed for the surfaces in the X-Y plane. The pre-crack is generated by deleting the atoms and bonds in the crack region in the X-Y plane throughout the Z-direction.

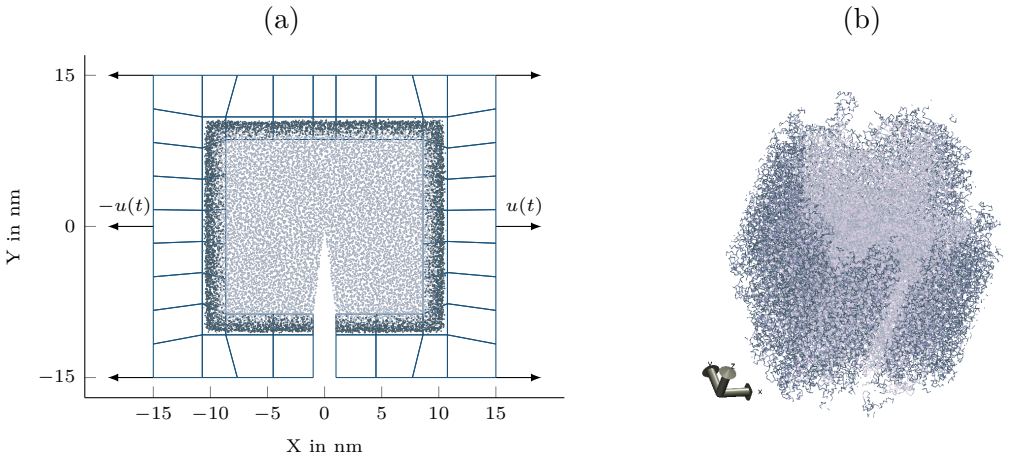

Figure 2: Illustration of (a) the coupled system and (b) the MD domain with a pre-crack. The MD system is generated with LAMMPS [14] and Figure (b) is visualized with VMD [8].

3 Numerical results

We study the mechanical behavior of a PS sample with different pre-cracks under tensile strain-controlled Mode-I loading in MD-FE coupling simulations of the systems demonstrated in Figure 2 (a).

3.1 Initial systems

The FE domain comprises 672 nodes and 378 trilinear hexahedral elements with $2\times2\times2$ Gauss points each. The detailed geometry of the FE domain on the X-Y plane is depicted in Figure 2 (a). The edge length of the coupled system in Z-direction is about 20.7nm and the pre-crack in the FE domain has a width of 2nm.

Same as in [13, 11, 23], the MD system is initially generated with PBC comprising 300 chains with 200 super atoms each, where the PBC on the surfaces in the X-Z and Y-Z plane are converted to SBC. Based on the MD system with hybrid boundary conditions, different pre-cracks as shown in Figure 3 are generated by deleting the corresponding atoms, bonds, and angles. These MD systems are denoted as 1, 2, and 3, respectively.

3.2 Relaxation

MD system 2 with a V-shaped crack and system 3 with a U-shaped crack are relaxed upon embedding into the FE domain with fixed FE boundaries in the Y-Z plane. To this end, the positions of the FE nodes not subjected to prescibed boundary conditions are

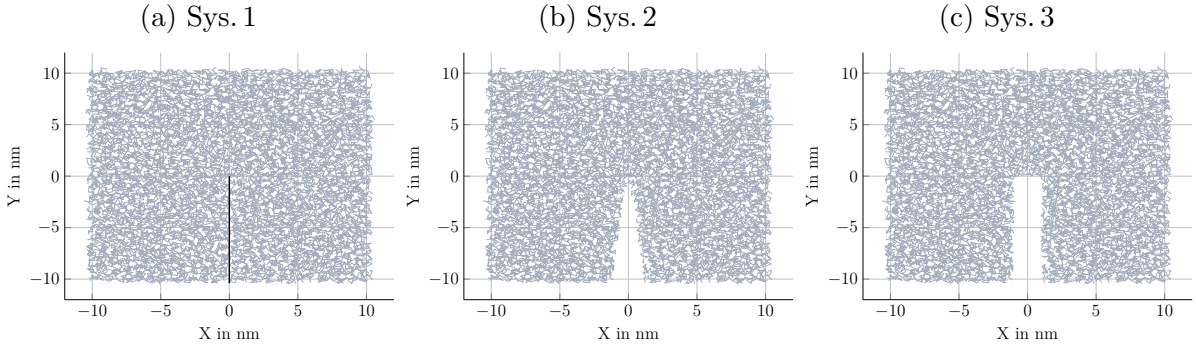

Figure 3: Initial configuration of the MD domain with different pre-cracks where the bonds crossing the plane represented by the black line are cut in Sys. 1 while the atoms in the corresponding crack areas are deleted in Sys. 2 and 3.

updated together with the anchor points during an MD-FE iteration step which comprises 10,000 MD timesteps with a size of $\Delta t^{MD} = 10$fs. Thus, each individual MD-FE iteration step corresponds to a time span of 0.1ns, which is repated 400 times to relax the MD domain within a total time of 40 ns.

The snapshots of the MD systems at the first and last two iterations at time $t = 0.1$ns, 0.2ns, 39.9ns, and 40ns are depicted in Figure 4, where a partial closing of the pre-crack can be observed. A much faster evolution of the crack occurs in the MD system with the V-shaped crack as obvious from comparing the crack shape at the first two iterations to its initial configuration in Figure 3. The comparison between the systems at the last two iterations demonstrates a convergence in terms of the crack shape. The closing of the crack during relaxation might be caused by the pair potential, which is in the same order of magnitude as the bond potential, such that the sharper pre-crack, i.e., with closer particles at the crack tip, exhibits faster dynamics. However, this shape-dependence of the crack relaxation is beyond the scope of this conceptual paper and thus not further studied here. Furthermore, the relaxed systems 2 and 3 at time $t = 40$ ns will serve as additional initial systems denoted as 4 and 5, respectively, in the following simulations under Mode-I loading conditions.

3.3 Mode-I Deformation

All coupled systems as shown in Figure 2 (a) are deformed under strain-controlled Mode-I loading conditions by prescribing the displacement of the FE surfaces in the Y-Z plane, whereas the other surfaces are subjected to free traction.

The overall loading strain in X-direction of the coupled system at time t is defined by

$$\tilde{\varepsilon}(t) = \frac{l_x(t) - l_x(t=0)}{l_x(t=0)} \tag{1}$$

with the edge length l_x of the FE domain in x-direction. The strain is applied step-wise as

$$\tilde{\varepsilon}(k\Delta t^{\text{ls}}) = \Delta\varepsilon^{\text{ls}} k \tag{2}$$

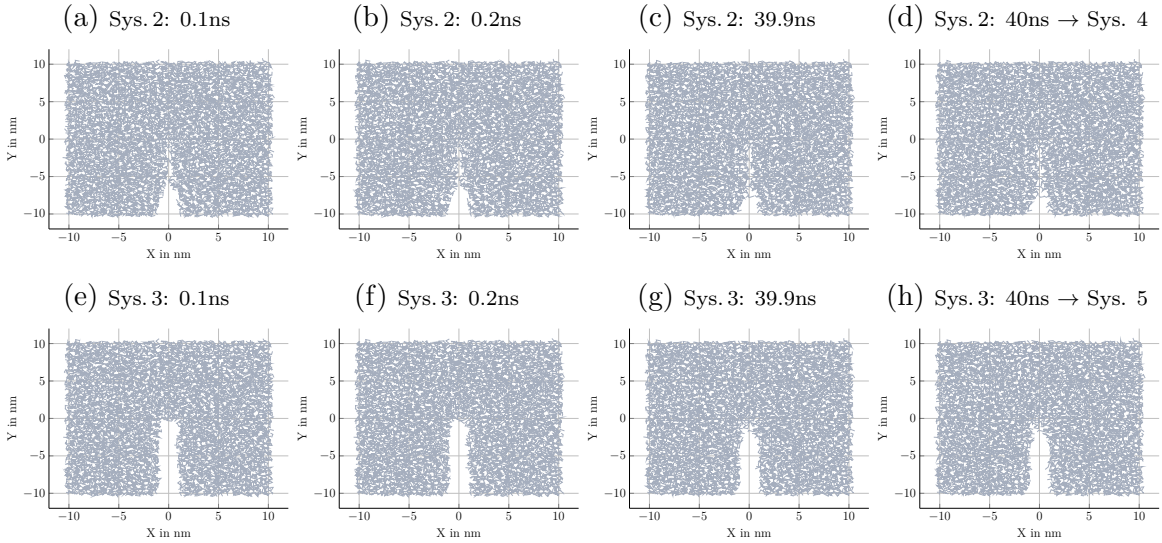

Figure 4: Snapshots of the systems Sys. 2 and Sys. 3 at different times during relaxation. The systems (d) and (h) are referenced as initial systems 4 and 5 in the following simulations.

with the load steps $k = 1, 2, ..., n^{\mathrm{ls}}$, the constant time step size Δt^{ls}, and the constant incremental strain $\Delta\varepsilon^{\mathrm{ls}}$ per load step. Consequently, the strain rate reads

$$\dot{\varepsilon} = \frac{\Delta\varepsilon^{\mathrm{ls}}}{\Delta t^{\mathrm{ls}}}. \tag{3}$$

Within each load step, the MD domain is run n^{MD} times with time step size Δt^{MD} such that $\Delta t^{\mathrm{ls}} = n^{\mathrm{MD}}\Delta t^{\mathrm{MD}}$.

The simulations are implemented with a constant strain rate of $\dot{\varepsilon} = 1\%/\mathrm{ns}$ up to an overall strain of 20%. Based on the parameter study of the Capriccio method for inelastic polymers [23] in terms of accuracy and computational cost, we choose the paremeters $\Delta\varepsilon^{\mathrm{ls}} = 0.005\%$ and $\Delta t^{\mathrm{MD}} = 5$ fs for our subsequent simulations. The other time-related parameters can be calculated based on their mutual dependencies as $\Delta t^{\mathrm{ls}} = 5\mathrm{ps}$ and $n^{\mathrm{MD}} = 1000$.

The deformation state of the MD domains for $\tilde{\varepsilon} = 5\%, 10\%, 15\%, 20\%$ are depicted in Figure 5. Compared to the other systems, the system 1 shows a minor change of the pre-crack, probably due the the dominant pair potentials, which prevents the separation of the crack surfaces. In contrast, the notch of the systems 2 – 5 propagates during deformation and the crack opens successively.

We further consider the stress distribution of the FE domain coupled with different MD systems. Since the stress in the bridging domain is weighted during the simulation, which is difficult to be accounted for in the post-processing, we only consider the stress on the outer surfaces. The Cauchy stresses σ_{xx} on the surfaces perpendicular to the X- and Y-directions at the last load step with the loading strain of 20% are plotted in Figure 6. On the loading surfaces, the deviations of the stress are very small. In contrast, the stresses on the front surface decrease, as expected, from the domain edge to the crack tip.

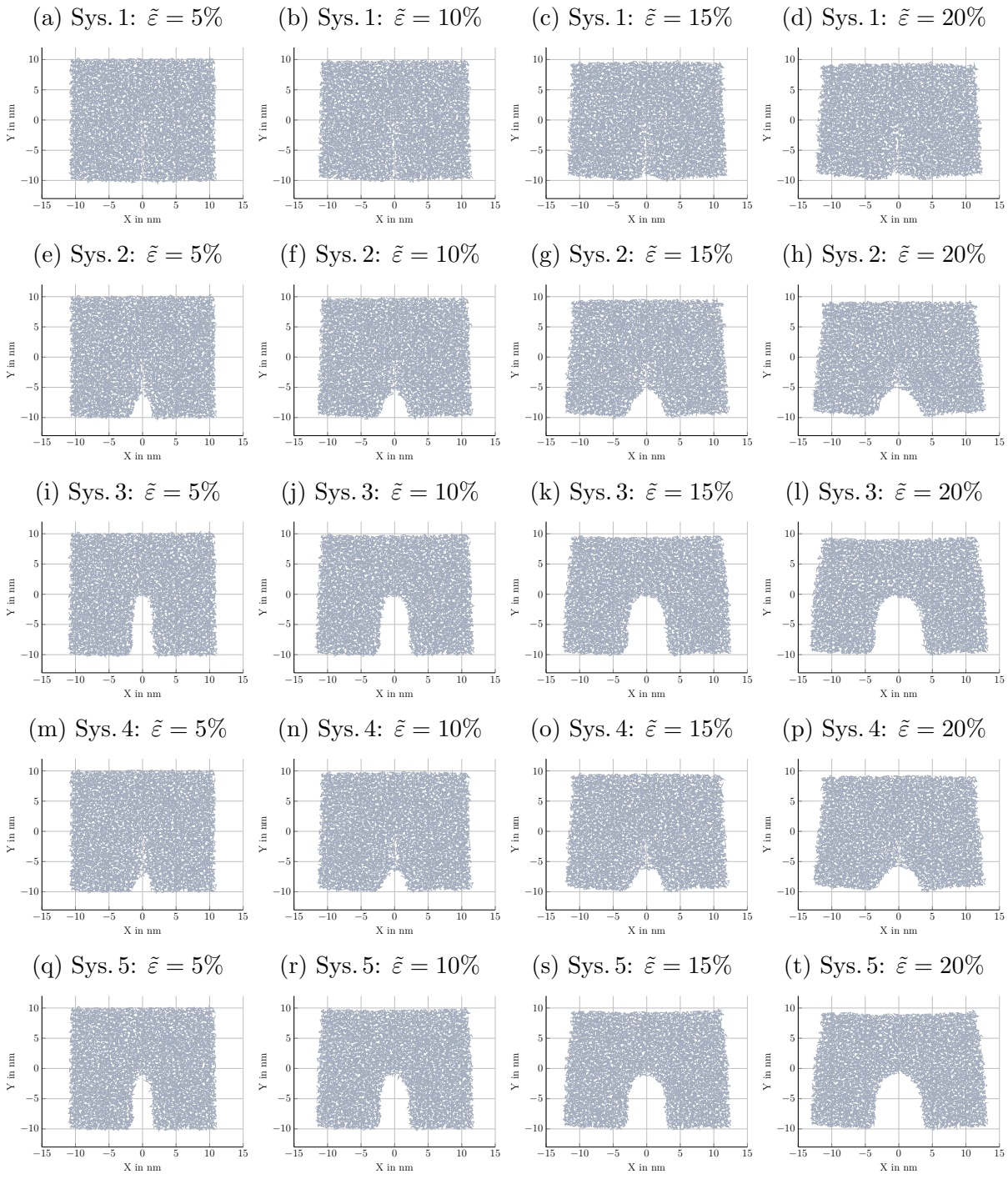

Figure 5: Deformation state of the MD systems 1 – 5 at different loading strain with constant strain rate 1%/ns.

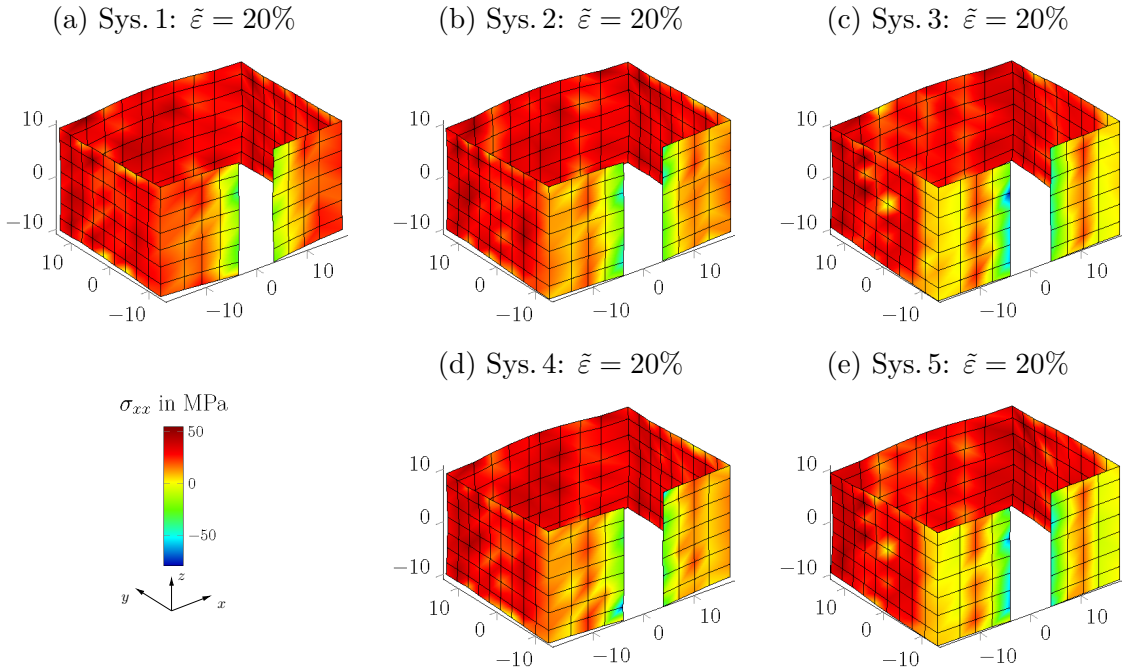

Figure 6: Stress distribution σ_{xx} on the outer surfaces of the coupled systems 1 – 5 at a loading strain of 20%.

Due to large deviations of the stress on the surfaces in X-Y plane, they are plotted separately in Figure 7 with a different colorbar for better readability. Here, only the systems 1 – 3 are considered with systems 4 and 5 rendering similar results compared to their original systems without relaxation. In Figure 7, stress concentrations can be observed near the corner and the crack tip, where actually finer elements are required. In addition, the systems with V- and U-shaped cracks show larger stress concentrations compared to system 1, which implies that larger pre-cracks lead to more pronounced inhomogeneities of the MD system.

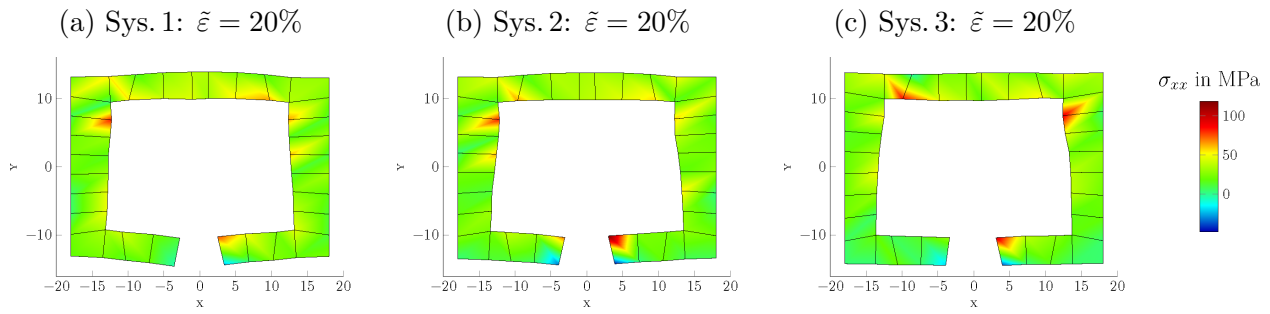

Figure 7: Stress distribution σ_{xx} on the outer surfaces of the FE domain coupled with the MD systems 1 – 5 at the loading strain of 20%.

The origin of these stress concentrations is not clear yet and requires further consid-

erations which are beyond the scope of this work. However, we assume that a significant contribution is due to the discrete nature of the MD domain, whereas further deviations from the ideal continuum case might be based on the current mesh size.

3.4 Loading with different strain rates

To demonstrate the versatility of the method, coupled simulations under Mode I loading conditions with different strain rates of 0.1%/ns and 20%/ns are implemented by varying the time step size during the calculation of the MD domain as $\Delta t^{\mathrm{MD}} = 50\mathrm{fs}$ and $\Delta t^{\mathrm{MD}} = 0.25\mathrm{fs}$, respectively, while keeping the other time-related parameters unchanged. The snapshots of system 5 at the strain of $5\%, 10\%, 15\%, 20\%$ with the different rates are depicted in Figure 8, where minor influence of the strain rate on the deformation of the MD domain can be observed. These results imply that larger deformation of the MD domain is required for the onset of the fracture at the crack tip, which will be considered in our future work.

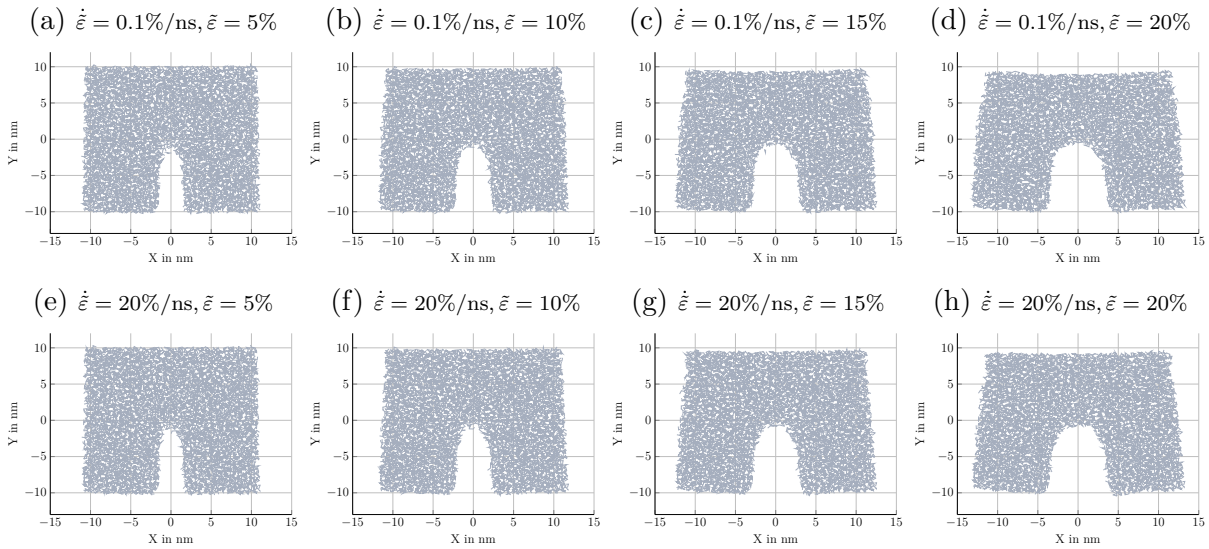

Figure 8: Deformation state of the MD system 5 at different loading strain with different constant strain rates of 0.1%/ns and 20%/ns.

4 Conclusion and outlook

In this paper, we apply an MD-FE partitioned-domain Capriccio method [23] to multiscale simulations of thermoplastic polymers under Mode-I loading conditions. The MD domain with different pre-cracks is loaded with different constant strain rates and we observe two opposite tendencies during relaxation and loading: First, during the initial relaxation process, the pre-defined cracks shrink with a clear dependency on the crack geometry. Second, duing the actual loading, the crack size increases with increasing strain. In the first case, we assume that the crack closing is caused by the rather strong non-bonded pair potentials. Without going into further details, such a behavior could lead to

an increased fracture resistance in case of cracks at the nanoscale. This, however, would require a thorough consideration of experimental results which cannot be provided here.

Although the multiscale method [23] shows good potential for studying fracture of polymers with rate-dependent properties, there are still several limitations in our current study. Firstly, a larger deformation state in the vicinity of the crack tip is necessary to observe crack propagation. This requires either an updated version of the constitutive model that can reproduce the mechanical behavior of the MD system under larger deformations, or a more appropriate geometrical setup of the FE domain such that larger deformations near the crack tip can be applied while keeping the strain of the FE domain small. Secondly, a finer FE mesh is required in the bridging domain to improve the accuracy of the calculations. Finally, a plausible bond breaking criterion should be considered in the MD system, where the process at the crack tip might have to be treated at finer resolution such as full atomistic resolutions or quantum mechanics. These aspects are currently work in progress.

Acknowledgment

This work is funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) project 396414850 “Identifikation von Interphaseneigenschaften in Nanokompositen” and project 377472739 “Research Training Group GRK 2423 Fracture across Scales – FRASCAL”.

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