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Q2 Determination of the size of the Representative Volume Element (RVE) for the simulation of heterogeneous polymers at finite strains

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ABSTRACT

The definition of the size of the Representative Volume Element (RVE) is extremely important for the mechanics and physics of heterogeneous materials since it should statistically represent the micro-structure of the material. In the present contribution, a methodology based on statistical analysis and numerical experiments is proposed to determine the size of the RVE for heterogeneous amorphous polymers subjected to finite deformations. The approach is applied to Rubber Toughened Polystyrene (RT-PS) composed by a two phase random micro-structure. Different micro-structural samples with two different percentages of rubbery particles, namely 10% and 15%, inside the micro-structure are studied. Periodic boundary conditions (PBC) are enforced to the RVE due to their fast convergence to the theoretical/effective solution when the RVE size increases. The Finite Element Method (FEM) is used in combination with mathematical homogenization to obtain the macro-stress. Two criteria are proposed for the RVE size determination. The proposed statistical-numerical approach is general and easy to use, when compared to the previously proposed approaches, and covers other criteria available in the literature.

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1. Introduction

Polymers have attracted a considerable attention from both academic and industrial communities due to their interesting characteristics such as good thermal and electrical insulation, lower density and also usually higher yield strain when compared to metals [1]. Besides the properties mentioned above, one of the relevant features of polymers is that complicated shapes can be easily fabricated through processes such as extrusion, injection molding and casting. For these reasons, polymers have a variety of applications, which are far more than any other class of materials, such as electronic products, automotive engineering, bio-mechanics, civil and building engineering, among others.

In spite of very interesting properties, polymeric materials also have some undesirable characteristics. For instance, glassy polymers show brittleness under specific conditions. Polystyrene (PS) and Polymethylmethacrylate (PMMA), among other glassy polymers, are considered brittle since they show brittle failure under low stress triaxiality such as uniaxial tension [2]. Therefore, in order to take advantage of the mentioned qualities of polymers and also improving the unfavourable characteristics, polymer

based composite materials have been the focus of a large number of studies during the last decade.

The most widely used strategy to improve the mechanical properties of materials consists in directly modifying the micro-structure of the material, see e.g. [3], which usually promotes a more heterogeneous nature including different phases in the micro-structure. Therefore, it becomes vital to establish relationships between the micro-structure of the material and the engendered macroscopic properties. There are different approaches to characterize the behaviour of materials with heterogeneous micro-structures. One possible approach is to fit material properties to experimental results for a continuum level constitutive model (e.g. [4]). Another approach consists in changing a continuum level constitutive model by including some information from the micro-structure of the material into the constitutive equations of the model so that the model could characterize the behaviour of the material with modified micro-structure (e.g. [2]). The most recent strategy entails the use of micro-mechanical or coupled multi-scale models based on homogenization techniques to transfer information between different scales (e.g. [5,6]). In fact, different homogenization techniques are used by different authors in order to model the mechanical behaviour of heterogeneous materials (see e.g. [7–10]). The reader is referred to [11] for a comprehensive review on homogenization techniques.

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In order to work within the framework of micro-mechanical and coupled multi-scale approaches, it is required to have the Representative Volume Element (RVE) of the heterogeneous material defined. In published literature, there are different definitions for the RVE (see e.g. [12–18]). What is common in the variety of definitions is that the RVE should accommodate enough information about the micro-structure, while its dimensions are smaller than the macroscopic dimensions, so that the concept of separation of scales is satisfied.

During the last two decades, there have been different methods employed by the scientific community to determine the RVE size (RVES) from image processing approaches (e.g. [19–21]), experimental-image processing methods (e.g. [22]), analytical approaches (e.g. [13,23]) and statistical-numerical methods (e.g. [24–29]). Grimal et al. [19] have derived the field of elastic coefficients from one acoustic microscopy image of a human femur cortical bone sample with an overall porosity of 8.5%. They have also used FEM to obtain the homogenized properties of RVES. Shan and Gokhale [20] have introduced a methodology involving image analysis techniques for determination of the RVES for Ceramic Matrix Composites (CMC) with different fiber sizes. Graham and Yang [21] determined the RVES for HY-100 steel using image analysis of the polished cross section of the material. Romero and Masad [22] have obtained the RVES for an asphalt concrete specimen utilizing an image analysis approach and they have verified their results with some mechanical tests. Drugan and Willis [13] employed a variational formulation to derive a nonlocal constitutive equation for a class of random linear elastic composite materials. They have estimated the RVES by comparing the magnitude of the non-local term and the portion of the equation that relates ensemble average stresses and strains through a constant "overall" modulus tensor. Sebsadji and Chouicha [23] have proposed an analytical approach, using fractal analysis, to obtain the RVES for concrete mixtures. Kanit et al. [24] have performed a statistical analysis of the numerical examples in order to quantify the RVES for two-phase three-dimensional Voronoi mosaic in the case of linear elasticity and thermal conductivity. Kanit et al. [25] have extended the statistical-numerical approach introduced in the previous work by Kanit et al. [24] to the case of real micro-structures of two materials from the food industry for RVES determination. Pelissou et al. [26] have started from the work of Kanit et al. [24] and introduced a new approach for RVES determination for a metal matrix composite with randomly distributed aligned brittle inclusions. Skarzynski and Tejchman [27] have used two different approaches, within a statistical analysis, to determine the RVES for softening quasi-brittle materials. The first approach, failure zone averaging approach, was previously introduced by Nguyen et al. [30]. Gitman et al. [28] investigated RVE existence in different stages of loading, elastic-hardening-softening, and also proposed a method to determine the RVES for random three-phase (matrix, inclusion and ITZ) heterogeneous materials. Stroeve et al. [29] have used different criteria (peak load, dissipated energy, strain concentration factor), within a statistical-numerical framework to determine RVES for materials with particles in a matrix material.

In this contribution, a statistical-numerical approach is proposed to determine the RVES for heterogeneous amorphous polymers subjected to finite deformations. Polystyrene (PS) is used as the matrix material toughened with rubbery particles. Two different volume fraction of inclusions are considered. The paper is structured as follows. Section 2 presents the kinematics of the micro-scale problem. The material model used for the polymeric matrix material is presented in Section 3. Section 4 presents some numerical examples at the micro-structural level with different percentage of inclusion volume fraction of rubbery particles. Section 5 details the proposed statistical-numerical approach for

determination of the RVES which is illustrated with numerical examples. Finally, Section 6 is devoted to some conclusions and concluding remarks.

2. Kinematics of the micro-scale problem

In this section, the mathematical formulation and main kinematical assumptions governing this study will be introduced. If $\mathbf{x}$ is an infinitesimal point at the macro-scale, $\mathbf{y}$ is an infinitesimal point on the RVE domain and $\mathbf{T}(\mathbf{y})$ is a generic field defined over the RVE domain, the homogenized response of the field, $\mathbf{T}$, for the macroscopic point to which the RVE is linked, is obtained by the following volume average relation:

$$\mathbf{T}(\mathbf{x}) = \frac{1}{V} \int_{\Omega} \mathbf{T}(\mathbf{y}) dV, \quad (1)$$

where V is the RVE volume and Ω is the RVE domain. It must be emphasized that the above relation can be applied to both undeformed and deformed configurations of the RVE. In this study, all the homogenized variables are defined as the volume average of the respective quantities over the initial configuration of the RVE.

The deformation of the RVE is driven by the macroscopic deformation gradient, which is the volume average of the microscopic deformation gradient:

$$\mathbf{F}(\mathbf{x}, t) = \frac{1}{V_0} \int_{\Omega} \mathbf{F}(\mathbf{y}, t) dV. \quad (2)$$

The microscopic deformation gradient is given by:

$$\mathbf{F}(\mathbf{y}, t) = \mathbf{I} + \nabla_p \mathbf{u}(\mathbf{y}, t), \quad (3)$$

where $\mathbf{I}$ is the second order identity tensor, $\mathbf{u}(\mathbf{y}, t)$ is the displacement field at micro-scale and $\nabla_p(*)$ is the material gradient operator. Using relations (2) and (3), the macroscopic deformation gradient is given by:

$$\mathbf{F}(\mathbf{x}, t) = \mathbf{I} + \frac{1}{V_0} \int_{\Omega} \nabla_p \mathbf{u}(\mathbf{y}, t) dV. \quad (4)$$

The displacement field at the micro-scale can be split into two terms:

$$\mathbf{u}(\mathbf{y}, t) = [\mathbf{F}(\mathbf{x}, t) - \mathbf{I}] \mathbf{Y} + \tilde{\mathbf{u}}(\mathbf{y}, t), \quad (5)$$

where the first term is the linear displacement varying linearly with $\mathbf{Y}$, which represents the RVE coordinates in the reference configuration, and the second term denoted by $\tilde{\mathbf{u}}(\mathbf{y}, t)$ is the displacement fluctuation field. Using Eqs. (3) and (5), the microscopic deformation gradient, $\mathbf{F}(\mathbf{y}, t)$, can be expressed as:

$$\mathbf{F}(\mathbf{y}, t) = \mathbf{F}(\mathbf{x}, t) + \nabla_p \tilde{\mathbf{u}}(\mathbf{y}, t). \quad (6)$$

Doing some algebraic manipulations, using divergence theorem and relations (2) and (6), results in the *minimal kinematical admissible constraint*:

$$\int_{\partial\Omega} \tilde{\mathbf{u}}(\mathbf{y}, t) \otimes \mathbf{N}(\mathbf{Y}) dA = 0, \quad (7)$$

where $\mathbf{N}(\mathbf{Y})$ denotes the outward unit vector to the undeformed boundary of the RVE. The virtual work equilibrium of the RVE states that:

$$\int_{\Omega} \mathbf{P}(\mathbf{y}, t) : \nabla \eta dV - \int_{\Omega} \mathbf{B}^{ref}(\mathbf{y}, t) \cdot \eta dV - \int_{\partial\Omega} \mathbf{T}^{ref}(\mathbf{y}, t) \cdot \eta dA = 0 \quad \forall \eta \in \nu, \quad (8)$$

where $\mathbf{B}^{ref}(\mathbf{y}, t)$ and $\mathbf{T}^{ref}(\mathbf{y}, t)$ are respectively the reference body force and the reference boundary traction force of the RVE. The symbol ν denotes the space of the virtual admissible displacement fluctuation field η . The first Piola–Kirchhoff stress of the infinitesimal point at the macro-scale is the volume average of

corresponding stress at the micro-scale:

$$\mathbf{P}(\mathbf{x}, t) = \frac{1}{V_0} \int_{\Omega} \mathbf{P}(\mathbf{y}, t) dV. \quad (9)$$

In order to relate the micro-scale and macro-scale, the *Hill–Mandel* principle is used. This principle is expressed as follows:

$$\mathbf{P}(\mathbf{x}, t) : \dot{\mathbf{F}}(\mathbf{x}, t) = \frac{1}{V_0} \int_{\Omega} \mathbf{P}(\mathbf{y}, t) : \dot{\mathbf{F}}(\mathbf{y}, t) dV, \quad (10)$$

which states equality between the macroscopic stress power and volume average of microscopic stress power over the RVE domain. The *Hill–Mandel* principle can be equivalently written as:

$$\int_{\partial\Omega} \mathbf{T}^{ref}(\mathbf{y}, t) \cdot \dot{\mathbf{u}}(\mathbf{y}, t) dA = 0, \quad (11)$$

$$\int_{\Omega} \mathbf{B}^{ref}(\mathbf{y}, t) \cdot \dot{\mathbf{u}}(\mathbf{y}, t) dV = 0. \quad (12)$$

2.1. Periodic boundary condition

The periodic boundary condition (PBC) is the most commonly adopted kinematical boundary constraint for the solution of RVE equilibrium problem. It has been demonstrated that PBC converges faster to the effective (or theoretical) as the RVE size increases [24,31]. In other words, for the same RVE size, PBC gives a better estimation of the effective properties in comparison to the other boundary conditions. This boundary condition is particularly suitable for the analysis of the mechanical behaviour of materials with periodic or even quasi-periodic microstructure. This boundary condition assumes that the boundary of the RVE is composed of negative and positive parts:

$$\partial\Omega = (\Gamma_i^- \cup \Gamma_i^+), \quad (13)$$

where each point $\mathbf{y}^+ \in \Gamma_i^+$ has a counterpart point $\mathbf{y}^- \in \Gamma_i^-$. Furthermore, the following condition must be satisfied:

$$\mathbf{n}^+ = -\mathbf{n}^-, \quad (14)$$

where $\mathbf{n}^+$ and $\mathbf{n}^-$ are the unit normal vectors of Γ_i^+ and Γ_i^- at points $\mathbf{y}^+$ and $\mathbf{y}^-$, respectively. The displacement fluctuation field for periodic boundary condition is periodic:

$$\dot{\mathbf{u}}(\mathbf{y}^+, t) = \dot{\mathbf{u}}(\mathbf{y}^-, t), \quad (15)$$

and the traction field is anti-periodic:

$$\mathbf{T}^{ref}(\mathbf{y}^+, t) = -\mathbf{T}^{ref}(\mathbf{y}^-, t). \quad (16)$$

Therefore, the application of PBC on the RVE consists in the enforcement of an identical fluctuation field for each pair of corresponding boundaries of the RVE. This forces the RVE to be discretized with a conform mesh. Nevertheless, in order to apply periodic boundary conditions on arbitrary finite element mesh discretization, several strategies have been proposed (e.g. [32–34]).

A comprehensive account of the numerical aspects required for the computational implementation of multi-scale constitutive models within a generic non-linear implicit finite element framework at finite strains can be found in [35], among others.

3. Material model

Over the last century, a considerable effort has been devoted to the development of new material models capable of reproducing experimentally observed data. It can be said that the initiation of the efforts to determine the behaviour of polymers dates back to 1930s. Eyring [36] proposed a molecular theory for the yield stress of amorphous polymers. Later, Mooney [37] proposed a strain energy function for rubber elastic materials. Haward and Tackray [38] developed a one dimensional constitutive model for glassy polymers and Boyce et al. [39] established a three dimensional version of Haward and Tackray model. Another constitutive approach that has been introduced to characterize the deformation behaviour of polymers is the generalized compressive Leonov model (currently known as EGP model). This model has been proposed by Baaijens [40] and extended by Tervoort et al. [41] and Govaert et al. [42]. In addition to the models developed to characterize the behaviour of polymers at continuum level, some authors have devoted their work to predict the deformation behaviour of polymers using molecular dynamics simulations (see e.g. [43,44]) or through multi-scale models from the molecular structure to the macro-scale (see e.g. [45]).

This section briefly describes the elasto-viscoplastic constitutive model which is used in this study. The total deformation gradient, $\mathbf{F}$, is multiplicatively composed of the elastic deformation gradient, $\mathbf{F}^e$, and the plastic deformation gradient, $\mathbf{F}^p$, [46]:

$$\mathbf{F} = \mathbf{F}^e \mathbf{F}^p. \quad (17)$$

It should be emphasized that the aforementioned decomposition of the deformation gradient has been mainly attributed to Lee [47] but recently Sadik and Yavari [48] showed that the decomposition was first introduced by Bilby et al. [46]. To measure the elastic deformations, the logarithmic (or natural) Eulerian (or spatial) strain is adopted. The plastic flow rule of the model is characterized by the generalized Eyring equation, which is given by:

$$\mathbf{d}^p = \dot{\gamma}^{eq} \frac{\partial \Psi}{\partial \boldsymbol{\tau}} = \dot{\gamma}^{eq} \mathbf{N}, \quad (18)$$

where $\dot{\gamma}^{eq}$ is the equivalent rate of shear strain, $\boldsymbol{\tau}$ is the Kirchhoff stress tensor, Ψ is the dissipation potential, $\mathbf{N}$ is the flow vector and $\mathbf{d}^p$ is the spatial plastic stretching tensor. The dissipation potential of the model is assumed to be given by:

$$\Psi = \sqrt{\frac{1}{2} \mathbf{s} : \mathbf{s}}. \quad (19)$$

Consequently, the flow vector is obtained as:

$$\mathbf{N} = \frac{\partial \Psi}{\partial \boldsymbol{\tau}} = \sqrt{\frac{1}{2} \frac{\mathbf{s}}{\|\mathbf{s}\|}}, \quad (20)$$

where $\|\mathbf{s}\|$ is the norm of $\mathbf{s}$ which is given by:

$$\|\mathbf{s}\| = \sqrt{\mathbf{s} : \mathbf{s}}. \quad (21)$$

Combining relations (18) and (20), the multi-dimensional plastic flow rule of the model is obtained as:

$$\mathbf{d}^p = \dot{\gamma}^{eq} \sqrt{\frac{1}{2} \frac{\mathbf{s}}{\|\mathbf{s}\|}}. \quad (22)$$

Table 1

Material properties for PS required by the constitutive model.

	E (MPa)	ν	ΔH (J/mol)	A_0 (s)	τ_0 (MPa)	D_∞	h	μ	R	H (MPa)
PS	3300	0.37	1.7E+5	1.11E−20	2.559	9	60	0.14	8.3143	11

Fig. 1. Two different samples with a size of 10 μm and volume fraction of inclusions equal to (a) 10% and (b) 15%.

Fig. 2. Stress-deformation of the samples shown in Fig. 1, solid line: 10% and dashed line: 15%.

The equivalent rate of shear strain is given by:

$$\dot{\gamma}^{eq} = \frac{1}{A} \sinh\left(\frac{\tau^{eq}}{\tau_0}\right), \quad (23)$$

where τ^{eq} is an equivalent stress defined by:

$$\tau^{eq} = \sqrt{\frac{1}{2} \text{tr}(\mathbf{s}\mathbf{s})} = \sqrt{\frac{1}{2} \mathbf{s} : \mathbf{s}}, \quad (24)$$

where $\mathbf{s}$ is the deviatoric stress. Using relation (23) and also Eq. (24), the plastic flow rule can be expressed in another form:

$$\mathbf{d}^p = \frac{1}{A} \sinh\left(\frac{\tau^{eq}}{\tau_0}\right) \left(\frac{\mathbf{s}}{2\tau^{eq}}\right), \quad (25)$$

or equivalently,

$$\mathbf{d}^p = \frac{\mathbf{s}}{2A \left[\frac{\tau^{eq}}{\sinh(\tau^{eq}/\tau_0)} \right]}, \quad (26)$$

which can be represented as:

$$\mathbf{d}^p = \frac{\mathbf{s}}{2\eta(\tau^{eq})}, \quad (27)$$

The viscosity function, $\eta(\tau^{eq})$, is given by:

$$\eta(\tau^{eq}) = A \left[\frac{\tau^{eq}}{\sinh(\tau^{eq}/\tau_0)} \right]. \quad (28)$$

The rate of equivalent plastic shear can be expressed in terms of multi-dimensional plastic flow rule:

$$\dot{\gamma}^{eq} = \sqrt{2\mathbf{d}^p : \mathbf{d}^p}. \quad (29)$$

The scalar parameter A is given by:

$$A = A_0 \exp \left[\frac{\Delta H}{RT} + \frac{\mu P}{\tau_0} - D \right], \quad (30)$$

where the scalar A_0 is a constant or pre-exponential factor involving the fundamental vibration energy; the parameter ΔH is the activation energy; the parameter R is the universal gas constant; the scalar T is the absolute temperature; the parameter μ is a pressure coefficient related to the shear activation volume, V , and the pressure activation volume, Ω , according to:

$$\mu = \frac{\Omega}{V}. \quad (31)$$

The deformation behaviour of polymers is also dependent on the hydrostatic pressure and thus the hydrostatic pressure should take part in the constitutive relations of the material model. In relation (30), P represents the total hydrostatic pressure of the analysis. The parameter D , in relation (30), is a softening parameter. The evolution of the softening parameter is modelled according to [49]:

$$\dot{D} = h \left(1 - \frac{D}{D_\infty} \right) \dot{\gamma}^p, \quad (32)$$

where D_∞ is the saturation value of the softening parameter and $\dot{\gamma}^p$ is an equivalent plastic strain rate defined by [42]:

$$\dot{\gamma}^p = \sqrt{2\mathbf{d}^p : \mathbf{d}^p}. \quad (33)$$

Using relations (24), (28)–(30) and (32) together with some algebraic manipulations results in the following viscosity function:

$$\eta = A_0 \exp \left[\frac{\Delta H}{RT} + \frac{\mu P}{\tau_0} - D_\infty + D_\infty \exp \left(\frac{-h\sqrt{3} \bar{\epsilon}^p}{\sqrt{2} D_\infty} \right) \right] \left[\frac{\tau^{eq}}{\sinh(\tau^{eq}/\tau_0)} \right], \quad (34)$$

where $\bar{\epsilon}^p$ is the accumulated plastic strain. The accumulated plastic strain rate is defined by:

$$\dot{\bar{\epsilon}}^p = \sqrt{\frac{2}{3} \dot{\mathbf{e}}^p : \dot{\mathbf{e}}^p}. \quad (35)$$

The total stress, $\boldsymbol{\tau}^{total}$, is additively composed of driving stress, $\boldsymbol{\tau}^{driving}$, and hardening stress $\boldsymbol{\tau}^{hardening}$:

$$\boldsymbol{\tau}^{total} = \boldsymbol{\tau}^{driving} + \boldsymbol{\tau}^{hardening}. \quad (36)$$

Fig. 3. Morphologies of the samples with size of 10 μm and volume fraction of inclusions equal to (a) 10% and (b) 15%.

Fig. 4. Stress-deformation of the samples shown in Fig. 3, solid line: 10% and dashed line: 15%.

Fig. 5. Stress-deformation of the samples shown in Figs. 1(a) and 3(a), both with inclusion volume fraction equal to 10%: blue line Fig. 1(a) and red line Fig. 3(a). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

In this model, the hardening stress is given as the deviatoric strain multiplied by a hardening modulus:

$$\tau^{\text{hardening}} = H \epsilon_d, \quad (37)$$

where ϵ_d gives the total deviatoric strain and H is the hardening modulus, which is one of the material parameters. The deviatoric strain is given by:

$$\epsilon_d = \mathbf{I}_d : \epsilon, \quad (38)$$

where $\mathbf{I}_d$ is the fourth order deviatoric identity tensor.

Fig. 6. Stress-deformation of the samples shown in Figs. 1(b) and 3(b), both with inclusion volume fraction equal to 15%: Fig. 1(b) and red line: Fig. 3(b). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

For a more elaborate discussion on the constitutive model and also the integration algorithm and finite element implementation, the reader is referred to [50].

4. Numerical examples

In this section, some numerical examples are performed on samples of rubber toughened Polystyrene composed by two phases: Polystyrene (PS) and rubber. The material properties for PS are taken from van Melick et al. [52] and are given in Table 1. It should be mentioned that due to the deformation induced cavitation early during deformation [2], the rubbery particles are modelled as voids. Two levels of inclusion volume fraction, namely 10% and 15%, are considered and the inclusions size range is assumed to be from 0.7 μm to 1 μm .

Remark 1. The rubbery particles are chosen to be within the mentioned size range due to the fact that the rubber toughening process is normally performed using sub-micron sized rubbery particles [2,51].

The first sample size is 10 μm . The samples are analysed with a non-linear finite element framework at finite strains which contains the constitutive model described in Section 3. The numerical approach employed to analyse each micro-structural sample consists in enforcing a macroscopic deformation gradient, $\mathbf{F}$, to the RVE and solving the micro-scale equilibrium problem and then

obtain the homogenized response of the stress field. The numerical simulations are performed under tensile loading conditions with the following deformation gradient:

$$\mathbf{F} = \begin{bmatrix} 1.15 & 0 \\ 0 & 1 \end{bmatrix}. \quad (39)$$

Fig. 1 shows two samples with dimension of $10 \mu\text{m}$ and volume fraction of inclusions equal to 10% and 15% that were randomly

Fig. 7. Flow chart for RVEs determination.

Fig. 8. Different realizations for inclusion volume fraction equal to 10% and different volume element sizes, (a): $10 \mu\text{m}$, (b): $15 \mu\text{m}$, (c): $20 \mu\text{m}$.

generated. The standard six-noded plane strain triangular element, with three Gauss integration points, is adopted for the spatial discretization of the samples. The mesh discretization for both cases is defined such that the relevant features of the geometry are well captured without an excessive level of refinement with PBC enforced to the relevant edges. Fig. 2 shows the stress deformation for both samples subjected to tensile loading under periodic boundary conditions. As expected, the increase in the inclusion volume fraction from 10% to 15% leads to a significant decrease of the material response quantified by $P_{11} - F_{11}$ curve. In order to analyse the impact of changing the morphology of the micro-structure on the overall deformation behaviour, one more sample of each inclusion volume fraction is generated. The samples are shown in Fig. 3. Fig. 4 depicts the stress-deformation graph for the samples shown in Fig. 3. In order to directly compare the difference between different realizations of the same inclusion volume fraction, the stress-deformation graph for samples of the size of $10 \mu\text{m}$ and inclusion volume fraction of 10%, shown in Figs. 1(a) and 3(a), is shown in Fig. 5. Similar to Fig. 5, the stress-deformation graph for samples of the size of $10 \mu\text{m}$ and inclusion volume fraction of 15%, depicted in Figs. 1(b) and 3(b), is presented in Fig. 6. As can be seen in Figs. 5 and 6, changing the morphology of the micro-structure clearly affects the overall behaviour of the micro-structural samples. Therefore, it is necessary to define what is the appropriate RVE size for each volume fraction of inclusions, such that the results obtained can be regarded as representative. To achieve this, a combined numerical-statistical analysis is proposed in Section 5, where two criteria are introduced for the determination of the RVE size.

5. RVE size

As mentioned before, working within the framework of micro-mechanical modelling and coupled multi-scale modelling approaches requires an appropriate definition of the RVE. An RVE should be large enough to accommodate enough information from the material micro-structure and at the same time, small enough to comply with the separation of scales. In this section, a statistical procedure combined with numerical simulations to determine the RVEs is presented. Again, Rubber Toughened Polystyrene (RT-PS) is considered for the numerical examples. Two different volume fraction of inclusions are considered namely 10% and 15%. The initial

Fig. 9. Different realizations for inclusion volume fraction equal to 10% and volume element size equal to 10 μm .

Fig. 10. Stress-deformation of the samples shown in Fig. 9: blue line: Fig. 9(a), orange line: Fig. 9(b), green line: Fig. 9(c), red line: Fig. 9(d), black line: Fig. 9(e). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

Fig. 11. Average stress-deformation of the samples shown in Fig. 9 with shaded bounds of standard deviation.

Table 2

The percentage of (standard deviation/average) for some representative values of deformation gradient for the samples shown in Fig. 9.

Deformation gradient, F_{11}	1.01	1.02	1.05	1.08	1.12	1.15
Standard deviation/average (%)	1.26	4.90	12.85	13.55	14.25	14.42

volume element size is assumed to be 10 μm and the inclusion size range is from 0.7 μm to 1 μm .

The procedure for RVES determination is as follows. First, different distributions of one volume element size and inclusion volume fraction are generated. Then, the samples are loaded and the stress-deformation graphs are extracted. Two criteria are introduced in order to determine the RVES:

1. The standard deviation of the deformation behaviour, which is represented by a specific deformation measure, should not be more than a predefined percentage of average deformation behaviour. In this work, 10% is considered for the variation of the first invariant of the first Piola–Kirchhof stress tensor, $I_1(\mathbf{P})$. The criterion is given by:

$$S_{I_1(\mathbf{P})} = \sqrt{\frac{1}{n-1} \sum_{i=1}^n (I_1^i(\mathbf{P}) - \bar{I}_1(\mathbf{P}))^2} \leq (0.1 \times \bar{I}_1(\mathbf{P})) \quad (40)$$

where $S_{I_1(\mathbf{P})}$ is the standard deviation, n is the number of samples, $I_1^i(\mathbf{P})$ is the i th value of the first invariant of the first Piola–Kirchhof stress tensor and $\bar{I}_1(\mathbf{P})$ is the mean value which is given by:

$$\bar{I}_1(\mathbf{P}) = \frac{\sum_{i=1}^n I_1^i(\mathbf{P})}{n} \quad (41)$$

Checking this criterion ensures that changing the realization, for the same sample size and same inclusion volume fraction, does not remarkably change the deformation behaviour.

2. The average behaviour of all the realisations of the same volume element size, k , denoted by $\bar{I}_1(\mathbf{P})^k$ is within a desirable predefined error with the average response of the next volume element size, $k+1$, denoted by $\bar{I}_1(\mathbf{P})^{k+1}$:

$$\text{Error}(\%) = \frac{|\bar{I}_1(\mathbf{P})^{k+1} - \bar{I}_1(\mathbf{P})^k|}{\bar{I}_1(\mathbf{P})^{k+1}} \leq 10\% \quad (42)$$

In this work, 10% is considered for the variation of the average first invariant of the first Piola–Kirchhof stress tensor of two consecutive volume element size. The second criterion checks if by increasing the size of the micro-structural sample, the deformation behaviour changes. If no considerable change is observed, while increasing the sample size, this criterion is satisfied.

The next stage is to check the first criterion to see whether it is satisfied. If the first criterion is satisfied, different distributions of the next volume element size with the same inclusion volume fraction are to be generated and analysed and the second criterion is checked. In case the second criterion is satisfied as well, the previous volume element size is RVES. If the first criterion is not satisfied, the next volume element size would be considered as the initial volume element size and the same process, as mentioned above, would be done. This procedure is represented in Fig. 7.

Remark 2. The procedure suggested is basically a method for determining a numerical RVE size for a given heterogeneous

Fig. 12. Different realizations for inclusion volume fraction equal to 10% and volume element size equal to 15 μm .

Fig. 13. Stress-deformation of the samples shown in Fig. 12: blue line: Fig. 12(a), orange line: Fig. 12(b), green line: Fig. 12(c), red line: Fig. 12(d), black line: Fig. 12(e). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

material when the macroscopic stress and strain measures converge.

5.1. RT-PS with 10% of rubbery particles

Fig. 8 shows three micro-structural samples with 10% of inclusion volume fraction and different sizes namely, 10 μm , 15 μm and 20 μm .

Remark 3. Preliminary simulations with micro-structural samples smaller than 10 μm revealed that the RVE size could not be smaller

Fig. 14. Average stress-deformation of the samples shown in Fig. 12 with shaded bounds of standard deviation.

Table 3

The percentage of (standard deviation/average) for some representative values of deformation gradient for samples shown in Fig. 12.

Deformation gradient, F_{11}	1,01	1,02	1,05	1,08	1,12	1,15
Standard deviation/average (%)	0.73	1.41	4.70	4.99	4.99	4.95

than 10 μm according to the introduced criteria. Thus, the initial volume element size to start the analysis is chosen to be 10 μm .

Five different morphologies for inclusion volume fraction equal to 10% and volume element size equal to 10 μm are shown in Fig. 9. For each volume size, a trial and error algorithm is employed

Fig. 15. Different realizations for inclusion volume fraction equal to 10% and volume element size equal to 20 μm .

Fig. 16. Stress-deformation of the samples shown in Fig. 15: blue line: Fig. 15(a), orange line: Fig. 15(b), green line: Fig. 15(c), red line: Fig. 15(d), black line: Fig. 15(e). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

Fig. 17. Average stress-deformation of the samples shown in Fig. 15 with shaded bounds of standard deviation.

to compute different radii of inclusions, within the range 0.7–1.0 μm , such that the desired inclusion volume fraction is achieved. This procedure attempted to mimic the process of rubber toughening, which is usually performed using sub-micron sized rubbery particles with different radii. Once the radii of all inclusions for each volume size and volume fraction are defined, five realizations are generated using a random sequential addition algorithm that places the same inclusions within the domain. This strategy guarantees that all realizations have exactly the same inclusion volume fraction.

Table 4

The percentage of (standard deviation/average) for some representative values of deformation gradient for samples shown in Fig. 15.

Deformation gradient, F_{11}	1,01	1,02	1,05	1,08	1,12	1,15
Standard deviation/average (%)	0.85	2.28	6.76	7.86	8.99	9.33

Remark 4. As pointed out by Kanit et al. [24], the RVE size could be associated with the defined precision and also the number of realizations. We considered five realizations for each inclusion

Fig. 18. Average stress-deformation of the samples shown in Figs. 12 and 15. Solid line: volume element size = 20 μm and dashed line: volume element size = 15 μm.

Fig. 19. Difference between the behaviour of samples shown in Figs. 12 and 15.

volume fraction and volume element size, as considered by other authors (see e.g. [28]). Apparently, by increasing the number of samples and also reducing the errors defined above (in the two criteria), more accurate RVE size will be obtained.

Fig. 10 shows stress-deformation curves for different realizations of samples with 10% of rubbery particles and volume element size equal to 10 μm. In order to proceed with the determination of the RVES, it is required to check whether or not the first criterion is satisfied. Fig. 11 shows the average response of the samples shown in Fig. 9 together with upper and lower bounds of standard deviation, defined respectively by:

$$\text{Upper Bound} = \overline{I_1(\mathbf{P})} + S_{I_1(\mathbf{P})}, \quad \text{Lower Bound} = \overline{I_1(\mathbf{P})} - S_{I_1(\mathbf{P})}. \quad (43)$$

In this study, if the standard deviation is within 10% of the average response, it is considered acceptable. In other words, if the standard deviation of the deformation behaviour, through the whole deformation, is less than 10% of the average deformation behaviour, the first criterion is satisfied. For the first series of samples with inclusion volume fraction equal to 10% and volume element size equal to 10 μm, the first criterion is not satisfied. This means that during the deformation process, the standard deviation is not always less than 10% of the average deformation behaviour. Table 2 gives the values of the percentage of the standard deviation to the mean value as a function of the deformation gradient at some representative deformation gradients. According to the proposed algorithm, shown in Fig. 7, the next step is to generate samples with the same inclusion volume fraction and a bigger size.

Five different morphologies for inclusion volume fraction equal to 10% and volume element size equal to 15 μm are shown in Fig. 12. The same loading is applied to these realizations and the stress-deformation curves are given in Fig. 13. Fig. 14 shows the average response of the samples depicted in Fig. 12 with the upper and lower bounds of standard deviation. As expected, making a comparison between Figs. 10 and 13 and also between Figs. 11 and 14 shows that by increasing the volume element size from 10 μm to 15 μm, the deformation behaviour of different realizations are closer to each other.

For the samples in Fig. 12, the first criterion is satisfied, i.e. the values of the standard deviation during deformation are less than 10% of the average behaviour. The values of the standard deviation/average versus deformation gradient are given in Table 3.

The next stage consists in generating bigger samples in order to proceed with the determination of the RVES. Five different realizations with volume element size equal to 20 μm are shown in Fig. 15. The respective stress-deformation curves are shown in Fig. 16. Again, it is required to check if the first criterion is satisfied for the samples with volume element size equal to 20 μm. The average response of the samples shown in Fig. 15 with the bounds of standard deviation are given in Fig. 17. Table 4 shows that the values of standard deviation are within the bounds defined, 10% of the average. Hence, the second criterion should be checked. In this study, 10% difference between the average behaviour of two volume element size is allowed. In other words, if the average behaviour of samples with 15 μm of volume element size, shown in Fig. 12, are within 10% difference to the average behaviour of samples with 20 μm of volume element size, the second criterion is satisfied too. Fig. 18 shows the average responses of both series of samples. In order to appreciate the evolution of the difference between the two averages, Fig. 19 shows the measure of the difference, i.e. error, between the average behaviour of samples shown in Figs. 12 and 15 versus deformation gradient. In Fig. 19, the error is computed by:

$$\text{Error}(\%) = \frac{|\overline{I_1(\mathbf{P})}^{20 \mu\text{m}} - \overline{I_1(\mathbf{P})}^{15 \mu\text{m}}|}{\overline{I_1(\mathbf{P})}^{20 \mu\text{m}}} \times 100. \quad (44)$$

According to the values of the average deformation behaviours of the two series of samples and also the defined criteria and algorithm, it can be concluded that for Rubber Toughened Polystyrene (RT-PS) with 10% of rubbery particles, the size of the Representative Volume Element (RVES) can be considered as 15 μm.

Remark 5. The rather time consuming and cumbersome RVE size determination process is due to the finite deformations. From Fig. 10, it can be obviously realized that within small deformation, still in the elastic regime, all realizations depict the same behaviour. It can also be perceived that even smaller volume element sizes could be satisfactory for small deformations.

As stated before, the rubbery particles are considered as voids in this study due to the internal cavitation of the particles during tensile deformation. Thus, the material considered here is, in fact, porous PS. It is known that porosity magnifies the post-yield softening of polymers. Considering the stress-deformation curves of the samples with different sizes, Figs. 10, 13 and 16, it is possible to conclude that the differences appear mainly after the elastic domain, during softening. Due to the aforementioned reasons, the RVE size determined is, most likely, a representative size for other loading conditions as well. Therefore, the same methodology could be used to define such size using eventually a more appropriate deformation measure.

Remark 6. In this study the effect of inclusion size range on the RVES is not studied. Stroeven et al. [29] investigated the effect of particle size distribution on the RVES. They concluded, considering peak load and dissipated energy as driving parameters, that changing particle sizes does not significantly change the RVES.

5.2. RT-PS with 15% of rubbery particles

The next volume fraction of rubbery particles to be considered is 15%. Fig. 20 shows five different realizations for 15% inclusion volume fraction and 10 μm volume element size. The respective stress-deformation curves are given in Fig. 21. In order to check the first criterion, if the standard deviation is within the 10% of the average response, Fig. 22 presents the average response of the samples given in Fig. 20 together with the bounds of standard deviation of the samples responses. Table 5 shows the percentage of standard deviation/average through the deformation. The criterion is checked and it is not satisfied. The next stage is to generate samples with the same inclusion volume fraction and a bigger size. Fig. 23 shows five different morphologies for 15% inclusion volume fraction and 15 μm volume element size. Fig. 24 depicts stress-deformation curves for different realizations of samples with 15% inclusion volume fraction and 15 μm volume element size. The average response of the samples presented in Fig. 23 with the bounds of standard deviation is given in Fig. 25. Table 6 gives the values of standard deviation/average through the deformation. The standard deviation of the behaviour of the samples is within the defined 10% of the average. Samples with volume element size equal to 20 μm and inclusion volume fraction equal to 15% are to be generated in order to check the first and second criterion. Five different morphologies for inclusion volume fraction equal to 15% and volume element size equal to 20 μm are shown in Fig. 26. Fig. 27 depicts the respective stress-deformation curves. Fig. 28 shows the average response of the samples presented in Fig. 26 with the bounds of standard deviation. Table 7 depicts the standard deviation/average for different values of deformation gradient. The first criterion is satisfied for this series of samples too. The second criterion should be checked. Fig. 29 depicts the average response of the samples shown in Figs. 23 and 26.

Fig. 30 shows the difference between the average behaviour of samples depicted in Figs. 23 and 26. Based on the defined factors for determination of the RVES and also considering the values of the average deformation behaviours of the two series of samples, the RVES for Rubber Toughened Polystyrene (RT-PS) with 15% of rubbery particles could be chosen as 15 μm .

Remark 7. In this work a finite strain elasto-viscoplastic constitutive model, described in Section 3, is employed to model the non-linear behaviour of Polystyrene (PS). Since the viscoplastic formulation includes rate effects, which implicitly introduce a characteristic length through the viscosity, it acts as a regularization method. Therefore, the phenomenon of localization of strains

in a narrow band, while the rest of the material experiences unloading, is attenuated and pathological dependence on mesh discretization is alleviated. To confirm this, some of the micro-structural samples in this study were spatially discretized with

Fig. 21. Stress-deformation of the samples shown in Fig. 20: blue line: Fig. 20(a), orange line: Fig. 20(b), green line: Fig. 20(c), red line: Fig. 20(d), black line: Fig. 20(e). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

Fig. 22. Average stress-deformation of the samples shown in Fig. 20 with shaded bounds of standard deviation.

Table 5

The percentage of (standard deviation/average) for some representative values of deformation gradient for samples shown in Fig. 20.

Deformation gradient, F_{11}	1,01	1,02	1,05	1,08	1,12	1,15
Standard deviation/average (%)	3.50	7.36	12.48	13.87	14.61	15.17

Fig. 20. Different realizations for inclusion volume fraction equal to 15% and volume element size equal to 10 μm .

Fig. 23. Different realizations for inclusion volume fraction equal to 15% and volume element size equal to 15 μm .

Fig. 24. Stress-deformation of the samples shown in Fig. 23: blue line: Fig. 23(a), orange line: Fig. 23(b), green line: Fig. 23(c), red line: Fig. 23(d), black line: Fig. 23 (e). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

Fig. 25. Average stress-deformation of the samples shown in Fig. 23 with shaded bounds of standard deviation.

Table 6

The percentage of (standard deviation/average) for some representative values of deformation gradient for samples shown in Fig. 23.

Deformation gradient, F_{11}	1,01	1,02	1,05	1,08	1,12	1,15
Standard deviation/average (%)	1.29	3.48	6.34	7.96	9.14	9.62

stress tensor) were obtained for both cases of RT-PS with 10% and 15% of rubbery particles.

It should be emphasized that the statistical-numerical approach for RVES determination, introduced in this study, has certain

finer finite element meshes and the difference in the results was negligible.

Remark 8. One may consider more intermediate dimensions, between the considered sizes in this study, to obtain a more precise RVE size.

Remark 9. The authors also applied the two criteria for RVES determination, introduced at the beginning of this section, to the first component of the first Piola–Kirchhof stress tensor (P_{11}), which is basically the stress component in the loading direction, and similar results (as obtained using the first invariant of the

Fig. 26. Different realizations for 15% inclusion volume fraction and 20 μm volume element size.

Fig. 27. Stress-deformation of the samples shown in Fig. 26: blue line: Fig. 26(a), orange line: Fig. 26(b), green line: Fig. 26(c), red line: Fig. 26(d), black line: Fig. 26(e). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

Fig. 28. Average stress-deformation of the samples shown in Fig. 26 with shaded bounds of standard deviation.

Table 7

The percentage of (standard deviation/average) for some representative values of deformation gradient for samples shown in Fig. 26.

Deformation gradient, F_{11}	1.01	1.02	1.05	1.08	1.12	1.15
Standard deviation/average (%)	1.46	2.33	3.06	3.79	4.56	6.27

6. Conclusions

In this paper, some numerical examples on Rubber Toughened Polystyrene (RT-PS), with two different levels of Inclusion Volume Fraction, were presented. Having different morphologies showed the necessity of determination of the RVE Size. Due to moderate

advantages compared to the previously introduced methods:

- Using this approach it is not required to investigate different regimes of the deformation, namely elastic, yield and softening, separately and thus it facilitates the statistical study.
- All possible criteria (e.g. slope of the graph in different regimes of the deformation, the strain at which the material yields and dissipated energy) are covered using this approach.

Based on the analysis conducted, we conclude that for materials which show a moderate softening response, an RVE size can be determined.

Fig. 29. Average stress-deformation of the samples shown in Figs. 12 and 15. Solid line: volume element size = 20 μm and dashed line: volume element size = 15 μm .

Fig. 30. Difference between the behaviour of samples shown in Figs. 23 and 26.

post-yield softening behaviour of the material, determination of a minimum size for the microstructural samples is of great importance. This fact was shown by increasing the size of the micro-structural samples and observing the difference. According to the factors, commonly used in the definition of the RVE, two criteria were introduced for determination of the RVES. It is worth emphasizing that in case of non-softening materials, determination of the RVES would be much easier due to the fact that softening behaviour causes localization of the deformation and this deformation localization greatly affects the overall deformation behaviour of the material. It was concluded that for both cases of inclusion volume fraction equal to 10% and 15%, the RVES could be considered 15 μm . In the future, expanding this statistical analysis with a larger number of samples for each volume element size may lead to further accurate results for the RVES.

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