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# On the account of a cohesive interface for modeling the behavior until break of highly filled elastomers

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ABSTRACT

The nonlinear behavior and failure of highly filled elastomers are significantly impacted by the volume fraction, the size and nature of fillers and the matrix stiffness. Original experimental data obtained on glass beads reinforced acrylates and on propellants allow illustrating and discussing the main effects generally observed. In order to better understand the effects of the microstructure and constitutive parameters on the behavior and failure of highly filled elastomers, a composite model, represented by a 2D periodic cell with randomly dispersed particles, with an account of a cohesive zone at the filler/matrix interface is used. Finite element simulations with finite strain provide insight on the stress–strain responses dependence to the model parameters and allow defining a failure criterion perceived by the appearance of a critical fibrillar microstructure.

## 1. Introduction

Solid propellants are made of polymer networks in the rubbery state filled with a very large amount of rigid oxidizer and metal fillers (volume fraction ranging between 50% and 90%). In an attempt to develop new materials, it is often desired to improve both strain and stress at failure. In order to comply with such a challenge it is necessary to understand the impact of the various material parameters on the propellant mechanical behavior. When narrowing our interest to the damage and failure, neglecting the viscoelasticity, of highly filled elastomers (discarding thermoplastic matrices) containing micrometric particles (eliminating nanosize particles like carbon-black or silica fillers), experimental data are scarce in the literature. One may cite the work of Vratsanos and Farris (1993a), reporting experimental data featuring the effect of the amount of fillers, the size of the fillers and the strength of the adhesion at the filler/matrix interface on the behavior and failure of glass bead reinforced polyurethane composites. Since damage at the filler/matrix interfaces, recognized as matrix debonding also named dewetting, seems to affect significantly the behavior of such composites, account for cohesive zones at the filler/matrix interfaces is often used to model the behavior of such materials following either a micromechanics approach or a finite element numerical approach. Micromechanics modeling is found in the case of linear material response and infinitesimal strain (Dvorak and Zhang, 2001; Tan et al., 2005; Nie and Basaran, 2005; Inglis et al., 2007; Tan et al., 2007; Ngo et al., 2010), and nonlinear hyperelastic matrix behavior for moderate amount of fillers (Brassart et al., 2009). The main limits of the micromechanics approach rest on the complications raised by the nonlinear behavior of the elastomer matrix, the large deformation that it may be submitted to, the very high volume fractions of fillers, and on the difficulty to define a local criterion for matrix failure that is sensitive to the field heterogeneities induced by the microstructure. Various finite element formulations have been proposed (Zhong and Knauss, 1997, 2000; Matouš and Geubelle, 2006; Matouš et al.,

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2007; Moraleda et al., 2009; Ngo et al., 2010). Early papers (Zhong and Knauss, 1997, 2000; Matouš and Geubelle, 2006; Matouš et al., 2007) focus on the numerical feasibility and the effect of cohesive zones on the composite stress–strain behavior. Zhong and Knauss (1997, 2000) show interest in the impact of the size of particles and of the interactions between particles for simple cells containing four particles arranged in a square manner. Moraleda et al. (2009) have proposed an interesting study on the impact of the strength and toughness of the cohesive zones but that lacks a discussion of the length scale parameters, which is essential when accounting for cohesive zones. Finally, Ngo et al. (2010) were also interested in the effects of the model and microstructure parameters but limited their study to the case of a single filler within a matrix of linear behavior undergoing infinitesimal strain.

In the current contribution, it is proposed to look at the general characteristics of the uniaxial behavior until break of highly filled elastomers at the light of existing and original experimental data, and to draw a qualitative comparison between the highlighted tendencies and those obtained by finite element simulations on periodic cells containing randomly distributed rigid particles in a hyperelastic matrix with a cohesive zone at each filler/matrix interface. A two dimensional numerical study of the microstructure and constitutive parameters is carried out to better recognize the key parameters that could enhance the strain and stress at break of such composites. Finally, criteria for composite failure are enunciated for simulations.

## 2. Experimental evidences on the monotonic behavior of highly filled elastomers

This section aims at reporting the effects of the material parameters on the uniaxial tensile stress–strain response of highly filled elastomers. Since experimental data are scarce, it was decided to present original experimental data that would help discussing the results of the literature.

### 2.1. Materials

### 2.1.1. Solid propellants

Solid propellants such as produced by Herakles groupe Safran were considered. In order to test the effect of the particle size, a plasticized elastomer was reinforced by explosive organic fillers called A with two different granulometries, either centered around 3 μm of diameter with very small scatter or centered around 26 μm of diameter with a wide scatter. Materials with 49% and 61% volume fractions of filler were prepared. In order to test the impact of the adhesion at the filler/matrix interface, another plasticized elastomer was mixed with either filler A or filler B, the latter being expected to enhance the polymer adhesion at its surface. The distribution of diameters for the filler B was similar to the second granulometry of filler A: centered around 26 μm with equally wide scatter. Finally, two matrices with significantly different behaviors were reinforced by the same amount and same type of filler (A) to study the effect of the matrix stiffness on the behavior of the composite. Tests were conducted at room temperature on 1 cm wide, 4 cm long and 0.5 cm thick dog-bone samples on a Zwick Z1.0 machine with a 1 kN load cell. For each material, five samples were tested.

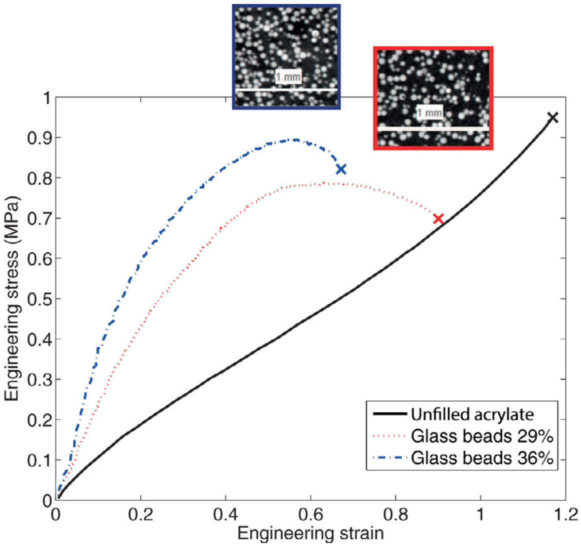

**Fig. 1.** Uniaxial tension stress–strain behavior of polyacrylate/glass beads composites with respect to the volume fraction of filler. Tomography images of the corresponding materials (insets) also illustrate the particle volume fraction.

### 2.1.2. Polyacrylate/glass beads composites

In order to avoid limiting ourselves to propellants, acrylate networks reinforced by micrometric glass beads were also prepared. Based on the tailorability of these networks (Safranski and Gall, 2008), the polymer network consists in a mix of 98 mol% Benzyl-methacrylate (BMA) monomer with 2 mol% Poly(ethylene glycol) dimethacrylate (PEGDMA) (550 g/mol) crosslinker copolymerized by UV reaction thanks to the 2,2-Dimethoxy-2phenylacetopeone photoinitiator. All products were used as received by Sigma-Aldrich. Sodocalcic glass beads with diameters in the range 45–63 μm were added as fillers. Final products are plates of 1.5 mm thickness after 55 min of curing within a UV chamber CL-1000. Dog-bone samples of 50 mm length and 4 mm width were punched from the plates and tested in uniaxial tension at 20 °C above the glass transition temperature in the rubbery state, on an Instron 5881 tensile machine equipped with an Instron thermal chamber and a 1 kN load cell.

For the acrylate composites as for the propellants, due to the large strain involved, the strain is measured locally with video extensometers during the tensile tests. Experimental tests were run at least three times for each material in order to assess the reproducibility of the experimental results that are presented below.

### 2.2. Effect of the filler volume fraction

The experimental data from Fig. 1 in Vratsanos and Farris (1993b) are often used as reference data describing the effect of the amount of fillers on the mechanical behavior of elastomers reinforced by spherical particles. Polyurethane composites containing from 0% to 50% of glass beads were tested by these authors in uniaxial tension while measuring the sample volume change. As expected, the initial modulus depends on the filler volume fraction. The onset of damage, detected by an increase of the sample volume, appears