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Experimental and finite element investigation of void nucleation in rubber-like materials

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ABSTRACT

This work deals with fracture of elastomers by cavitation. So, tension tests were achieved on specific volumetric specimens, the shape of which should induce void nucleation in the bulk of the material. Macroscopic behaviour of this material was related to the cavitation phenomenon. It was particularly proved that the slope break of the stress–strain curves coincides with the apparition of voids. Then, all experimental tests were numerically modelled using Finite Element Method (FEM) and results were analysed. Numerical study highlighted, among others, effects of specimen shape factor and those of filler volume fraction on the nucleation and growth of cavities.

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1. Introduction

Because rubbers are quasi-incompressible materials, microvoids can nucleate in their bulk under certain loading conditions. This cavitation phenomenon was early observed by Busse (1938), Yezley (1939). They observed cavities in the centre of so-called “pancake” specimens under tension loading. These specimens consist of thin rubber discs adhered to metal plates. Later, many authors investigated this phenomenon both experimentally and theoretically. Thus, Gent and Lindley (1959) analysed cavities nucleation in carbon-black-filled natural rubber (NR) using “poker chip” tests consisting in submitting pancake specimens to extension in the direction of the principal axis perpendicular to their larger surface. They showed that the initiation of cavities, due to a high stress triaxiality in the material, leads to a slope break of load–displacement curves. Using acoustic emission, Kakavas and Chang (1991, 1992) have also pointed out the nucleation of cavities in pancake samples submitted to traction loading.

Assuming a pure hydrostatic loading, the pioneering theoretical analysis of Ball (1982) led to a mathematical expression allowing the prediction of the cavity nucleation in a material. According to this theoretical approach, some authors also expressed such prediction with strain energy density functions (Stuart, 1985; Chou-Wang and Horgan, 1989; Horgan and Pence, 1989; Polignone and Horgan, 1993a; Polignone and Horgan, 1993b; Horgan and Polignone, 1995). Comparing the experimental results obtained

by Gent and Lindley (1959), and by Oberth and Bruenner (1965) with those issued from finite element method (FEM), Stringfellow and Abeyaratne (1989) confirmed that the theoretical criterion obtained by Ball (1982) provides a good estimation of the cavitation stress under a pure hydrostatic loading. A few years later, considering a neo-Hookean material, Hou and Abeyaratne (1992) extended for any 3D loading the cavity nucleation criterion proposed by Ball (1982). In fact, they drew in the three-dimensional principal stresses space a threshold surface delimiting the domain of the material safety. The pertinence of this approach was validated, on the one hand, by Chang et al. (1993) via FEM and, on the other hand, by Ganghoffer and Schultz (1995) using an asymptotic method. Diani (1999) completed this study by introducing a procedure allowing the description of the evolution of a pre-existing cavity during the loading.

When incompressibility of the material is not ensured, it is generally quite difficult to determine analytical solutions giving parameters governing the cavity nucleation. Nevertheless many authors showed that, beyond a certain critical value of the deformation, the cavitation solution is more stable (Ball, 1982; Horgan, 1992; Horgan, 1995; Murphy and Biwa, 1997; Xin-Chun and Chang-Jun, 2001). Recently, Kakavas (2002) studied the effect of the microvoids volume fraction on the stress–strain fields of compressible materials under monotonic loading.

Damage of rubber-like materials by cavitation was also analysed under cyclic loading (fatigue) by Dorfmann and Burtscher (2000), Dorfmann et al. (2002), Legorju-jago and Bathias (2002).

Moreover, void nucleation in rubber particles dispersed in a PMMA matrix was studied by Lazzeri and Bucknall (1993), Fond et al. (1996).

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The cavitation phenomenon in elastomers is an interesting but a complex problem because it requires advanced and up-to-date experimental facilities to detect the nucleation and to quantitatively measure the cavity growth. From an analytical point of view, it requires strong background to treat all the difficulties such as, among others, finite strains, incompressibility/compressibility and bifurcation of solutions.

The purpose of this work is to analyse the fracture of elastomers by cavitation process. More precisely, we first achieved specific experimental tests inducing voids nucleation in the bulk of the material and the obtained results have been analysed. These tests are then numerically modelled by FEM, and the effects of specimen shape factor and those of filler volume fraction on the cavitation are highlighted.

2. Theoretical background: analytical modelling of the cavitation phenomenon in rubbers

Many investigations dealing with the mechanical modelling of nucleation and/or growth of voids in rubbers were developed, by analysing the strain and stress fields on a Representative Volume Element (RVE) subjected to a certain loading. Depending on whether incompressibility is assumed or not, different approaches were proposed. In this section, only the analytical model developed by Ball (1982) and used in this study is presented.

Ball (1982) considered a spherical RVE of an incompressible, isotropic and hyperelastic material, with initial radius b_0 , subjected to a uniform radial pressure p on its external surface as illustrated by Fig. 1. In this figure, a and b represent the radii of the nucleated cavity and the RVE in the deformed state, respectively.

Considering a neo-Hookean mechanical behaviour, the analysis of the strain–stress field led Ball (1982) to the following expression of the critical hydrostatic pressure p_c , at the onset of cavity nucleation, in the center of the RVE:

$$p_c = \frac{\sigma_1 + \sigma_2 + \sigma_3}{3} = \frac{5\mu}{2}, \tag{1}$$

where σ_1 , σ_2 and σ_3 are the Cauchy stresses in the principal directions 1, 2 and 3, respectively, and μ is the material shear modulus.

Under incompressibility state, the Young’s modulus E is related to the shear modulus μ by $E = 3\mu$. Thus, the critical hydrostatic pressure p_c given by Eq. (1) can be rewritten as follows:

$$p_c = \frac{\sigma_1 + \sigma_2 + \sigma_3}{3} = \frac{5E}{6}, \tag{2}$$

which is in agreement with results found by Gent and Lindley (1959), Oberth and Bruenner (1965), Oberth (1967). Cho and Gent (1988) developed original experimental tests on a transparent silicone and found a critical pressure value in agreement with that

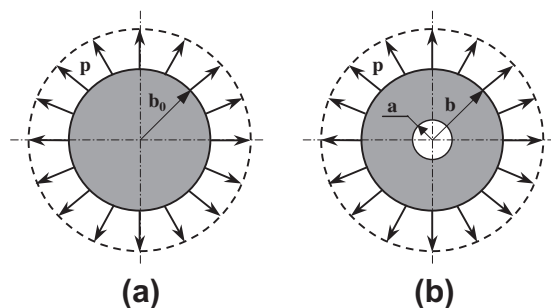


Fig. 1. Schematic representation of cavity nucleation with spherical model: (a) initial and (b) deformed configurations.

given by Eq. (2). Stringfellow and Abeyaratne (1989) also confirmed this result using FEM.

One of the most important restrictions of the Ball’s model is the assumption of a uniform external radial pressure on a spherical RVE while the loading is generally not purely hydrostatic.

Some authors tackled, either experimentally or numerically by using FEM, the cavity growth process (Williams and Schapery, 1965; Lindsey, 1967; Gent and Tompkins, 1969; Gent, 1990; Gent and Wang, 1991; Diani, 1999, 2001; Chang and Pan, 2001). Moreover, several authors analytically analysed the cavitation phenomenon in the case of compressible materials (Ertan, 1988; Horgan, 1992, 1995; Biwa, 1995; Murphy and Biwa, 1997; Xin-Chun and Chang-Jun, 2001). They mainly concluded that it is quite difficult to get pertinent solutions describing nucleation and/or growth of cavitations.

3. Experimental study

Hydrostatic depression tests were carried out on a carbon-black-filled styrene-butadiene rubber (SBR) vulcanizate. They consist in pulling out thin discs of this material in the direction perpendicular to their larger surface. Specimens were obtained by directly bonding, via a specific process developed and widely used in the rubber industry, the elastomer component to metal plates during the vulcanisation. These specimens were tested right after their elaboration. Because of the particular shape of these so-called pancake specimens, dimensions of which are summarised in Table 1, high stress triaxiality (hydrostatic stress state) is generated in the bulk of the material, increasing the hydrostatic pressure, which therefore favours the nucleation of cavities. In fact, Busse (1938) already observed voids nucleation in the center of similar specimens of rubber loaded in tension. These observations were also confirmed by Yerzley (1939) on a neoprene synthetic elastomer, Gent and Lindley (1959) on NR vulcanizate and Pond (1993) on filled NR.

Experimental data were recorded up to total breaking of the specimen and results are reported in Fig. 2 in terms of load as function of applied displacement. All curves exhibit linear evolution for small displacement range. Beyond a critical displacement, the material stiffness decreases and the curves show a non-linear evolution until total rupture of the specimen. The macroscopic slope change may be seen as an indicator of voids nucleation, which volume reaches a sufficient critical value allowing the modification in the global behaviour of the material. This is confirmed by the fracture surfaces shown in Fig. 3 exhibiting void footprint suggesting that the total rupture of the specimen happened by the process of nucleation and growth of cavities. Fracture surfaces also reveal that mechanisms of voids nucleation seem to depend on the specimen shape factor defined in this work as the loaded area to unloaded one (Gent and Lindley, 1959), i.e. $s = \pi R^2 / 2\pi Rh = D/4h = R/2h$ where R and h are the radius and the height of the specimen, respectively. It must be noted that this aspect ratio is sometimes defined in the literature as the unloaded area to loaded one, i.e. $s = 2\pi Rh / \pi R^2 = 4h/D = 2h/R$. However, our choice does not affect at all the analyses and the understanding of results reported in the present paper.

Note that the shape factor plays an important role in the nucleation and growth of voids. In fact, as shown in Fig. 3, the highest the shape factor, the highest the density of voids and the smallest

Table 1
Dimensions of pancake specimens.

Diameter (mm)	Height (mm)
100	2.5, 5, 10, 20, 25, 50

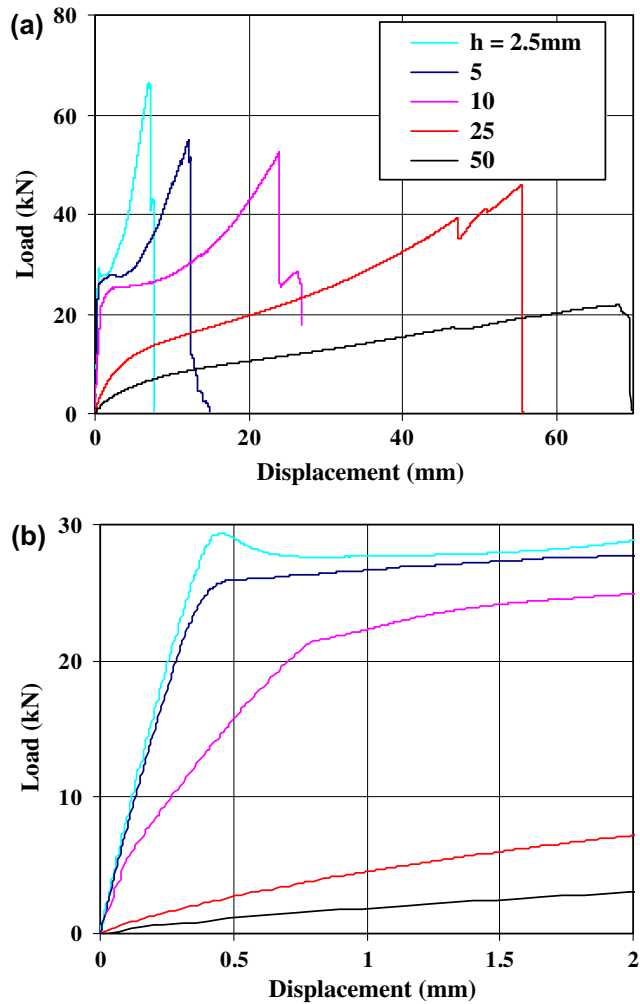


Fig. 2. Load–displacement curves of pancake specimens: (a) until total breaking (b) zoom at small strains.

their size. Below a certain value of the shape factor (here when $h \geq 25$ mm), the fracture process is not controlled by nucleation and growth of cavities but by initiation and propagation of a crack. That may explain why the load–displacement curves reported in Fig. 2, show that for high shape factor values, the slope change is well marked. However, for small values of s , this threshold is much less visible. Similar results were already found by Gent and Lindley (1959), Pond (1993) who reported that the slope break is frequently accompanied by an audible sound that is more magnified when increasing the sample shape factor. This is probably a consequence of the stress state increasingly close to a pure hydrostatic state as s increases. Fig. 2 also shows that the applied loads and displacements at the slope break depend on the specimen shape

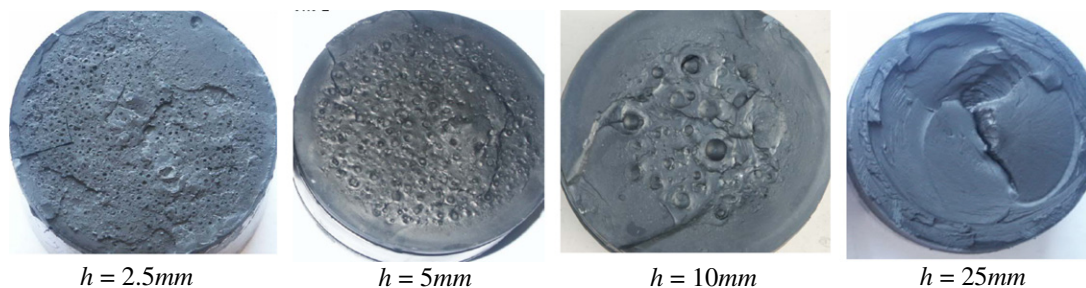


Fig. 3. Fracture surfaces of pancake specimens.

factor s . In fact, the critical load increases and the critical displacement decreases with s , but they both remain quite smaller compared to the total breaking values.

To put experimentally in evidence the coincidence of the slope change in the load–displacement curves with critical nucleation of internal flaws, Gent and Lindley (1959) and, then, Lindsey (1967) used a direct observation on a transparent rubber. Recently, Kakavas and Chang (1991, 1992) used an acoustic emission technique to analyse cavity nucleation in bonded unfilled nitrile rubber discs.

In the present work, to verify if the decrease of the apparent sample stiffness is due to the voids nucleation, the material volume variations were measured during extension of pancake specimens. A sudden change of material volume could be a signature of cavity nucleation in this material. The tests consist in extending such specimen in a sealed chamber filled with a quasi-incompressible liquid (Fig. 4). The void volume fraction appeared in the bulk of the tested elastomer could correspond to the liquid volume variation measured via calibrated vertical capillary tube.

Fig. 5 shows an example of obtained results in terms of load and volume variations as a function of imposed displacements for a specimen of 10 mm height. This figure clearly shows that, for small deformations, the material volume remains constant with some fluctuations related to the lack of accuracy of the experimental set-up. Then, it suddenly increases beyond a critical displacement, continuously and linearly as a function of the displacement up to total fracture of the specimen. The threshold point is located in the zone of slope break of load–displacement curves. Viscosity should maybe enhance the peak of this slope break. Similar observations were made for all specimens with height less than 25 mm. For specimens with $h \geq 25$ mm, no sudden volume change was noted confirming that below a certain value of the shape factor, the fracture process is not based upon cavitation because the stress state in the centre of the specimen moves away from a pure hydrostatic loading. As a consequence, one can conclude that a sudden change of material volume is a signature of voids nucleation. Therefore, the stiffness change observed in the load–displacement curves corresponds to a damage process induced by initiation of micro-voids that grow when increasing the loading up to total rupture of the specimen by coalescence of these voids.

4. Numerical modelling

The aim of this FE study is to verify the pertinence of the above experimental results interpretations and to further analyse the damage of rubbers by cavitation. Even a complete study requires investigating void growth process and damage accumulation, we only focus in this work on the void nucleation prediction in rubbers. So, pancake specimens experimentally tested were numerically simulated under tension loading, using the FE program “Marc”. Cylindrical RVE containing a rigid inclusion was also modelled.

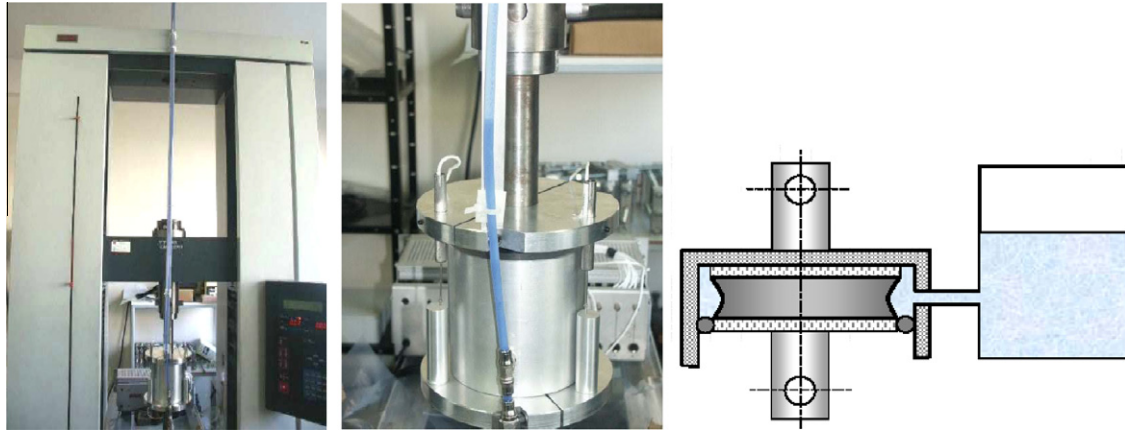


Fig. 4. Experimental set-up allowing the evaluation of elastomer volume variations.

4.1. Effects of specimen shape factor on the cavitation

Poker chip tests were modelled using axisymmetric 2D-model and considering a neo-Hookean constitutive law given by the following equation:

$$W = C_{01}(I_1 - 3), \tag{3}$$

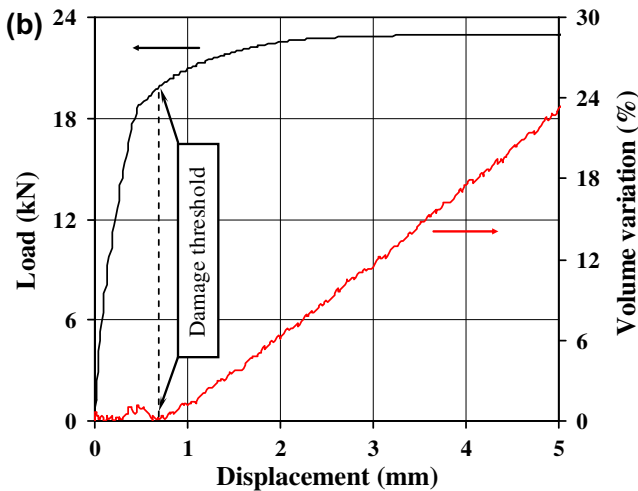
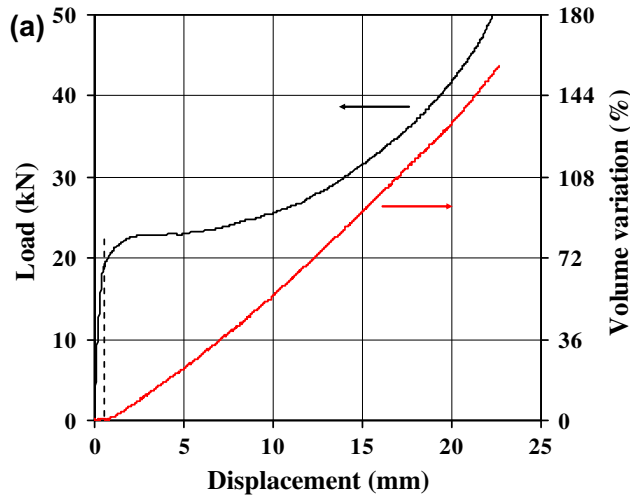


Fig. 5. Example of loads and specimen volume variations as a function of the imposed displacements ($h=10$ mm): (a) until total breaking (b) zoom at small strains.

where W is the strain energy density, C_{01} is the material constant and I_1 is the first invariant of the right Cauchy-Green strain tensor. The bulk factor or the incompressibility factor K of the material was measured via oedometric test and found equal to 2200 MPa, suggesting that the incompressibility assumption is reasonable. In this case, the constant C_{01} is directly related to the Young's modulus E by $C_{01} = E/6$. The value of E was provided by dynamical tests achieved on a DMA machine under small deformation range and it was found equal to 5 MPa. Metal plates on which rubber specimens were bonded were assumed to be infinitely rigid. Only the elastomeric specimen was modelled and it was meshed using four nodes quadrilateral elements. The meshing was refined in the vicinity of the central zone of the specimen, where the hydrostatic pressure gradient is expected to be important. The FE calculation was achieved by gradually increasing the displacements applied to the nodes located at the top of the sample. Stress and strain fields were evaluated on the whole specimen and the hydrostatic pressure was therefore available. Fig. 6 shows the distribution of the hydrostatic pressure in the samples for the different values of the shape factor and for displacements close to the expected cavity initiation threshold corresponding to the sudden volume change of the material (Fig. 5). Fig. 6 clearly highlights that the shape factor influence is predominant in the distribution. In fact, for high values of this parameter, the hydrostatic pressure is greatest in the central region of the specimen, while, when s decreases, the maximum pressure moves towards the interface.

Finally, the critical global strain ϵ_c corresponding to the onset of void nucleation was evaluated and it appeared strongly dependent on the specimen shape factor s , as clearly pointed out in Fig. 7. In fact, ϵ_c decreases when increasing the factor s , suggesting that the more confined the specimen, the smaller the strain is at breaking.

Another interesting aspect that deserves consideration is the effect of filler volume fraction on cavitation phenomenon in rubbers. So, we focused a part of our study on this aspect that is not largely tackled in the literature. The obtained results are presented and analysed in the next section.

4.2. Effects of filler volume fraction on the cavitation

To highlight the cavitation phenomenon in rubbers, some authors carried out experimental tests on cylindrical samples of transparent elastomer in which a rigid spherical particle made of glass or steel is embedded (Oberth and Bruenner, 1965; Gent and Park, 1984). These specimens are then submitted to a uniaxial loading. The effects of the elastomer matrix Young's modulus were investigated by varying, for a given material, the rate of cross-linking

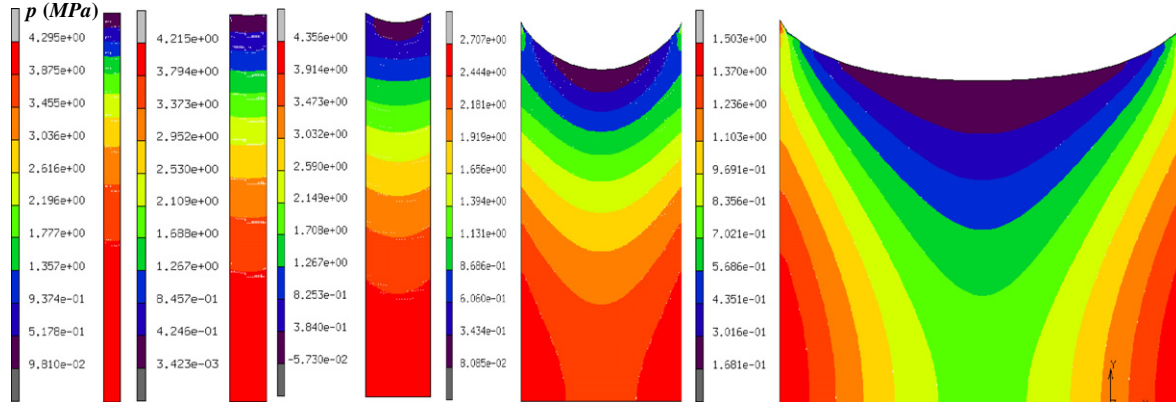


Fig. 6. Influence of the specimen shape factor on the distribution of the critical hydrostatic pressure p_c (MPa) evaluated for displacements close to the expected cavity initiation threshold.

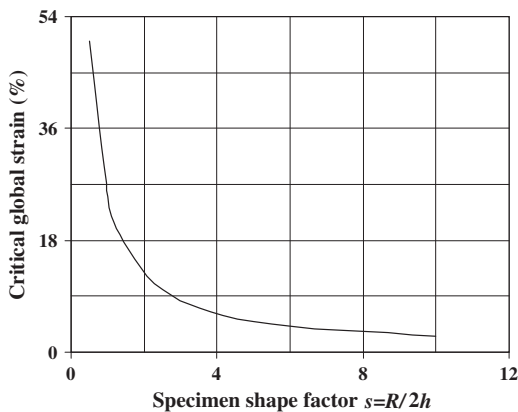


Fig. 7. Critical global strain at the onset of void nucleation as a function of the specimen shape factor.

during the vulcanisation process. They showed that cavities first nucleate at the pole of the inclusion suggesting that higher stress triaxiality is involved in this region.

In this study, we modelled by FEM, using “Marc” software, a cylindrical RVE containing a rigid spherical particle in its centre and submitted to uniaxial tension in the direction of its long axis. This specimen is 30 mm long and its diameter is 12 mm. Several values of reinforcing particle radius were considered, $r(\text{mm}) = \{0.5, 1, 2, 3, 4, 4.5\}$, corresponding to the filler volume fractions f (filler volume/composite volume $\times 100$) = $\{0.015, 0.12, 1, 3.5, 7.5, 10\}$. This particle was supposed infinitely rigid and perfectly adhered to the matrix. A 2D axisymmetric model was selected and the matrix behaviour is assumed to obey to a neo-Hookean law (Eq. (3)) with Young’s modulus $E = 5$ MPa. The material is again supposed to be incompressible since its bulk factor K , experimentally measured via oedometric tests, was found equal to 2200 MPa. Because of symmetries, only a quarter of the specimen was analysed.

Selected grid work contains exclusively four node quadrilateral elements and it is refined in the vicinity of the reinforcing particle. As for pancake specimens (Section 4.1), the FE calculation was achieved by gradually incrementing the displacements applied to the nodes situated on an extremity side of the cylinder. The quantity “ $p - 5E/6$ ” corresponding to the gap between the actual pressure and the critical pressure value according to the Ball’s criterion, was evaluated on the whole sample. Fig. 8 shows, as an example for particles of radius $r(\text{mm}) = \{0.5, 2, 4.5\}$, a map of the distribution of this quantity in the specimen for a given

displacement. Positive values indicate that the pressure given by the Ball’s criterion has been exceeded, while negative values correspond to region where the criterion is not yet reached. According to the Ball’s criterion, it is clearly pointed out that void nucleation is located at the filler poles which is in agreement with the experimental observations of Oberth and Bruenner (1965), Gent and Park (1984).

However, it must be noted that it is more complicated to model the phenomenon of cavitation when considering, as it is the case in practice, several particles in the matrix, because it is indispensable to take interaction effects between fillers into account. Homogenisation methods or, generally, multiscale approaches should be strong alternative techniques that could bring out some interesting responses in such case.

To further analyse numerical results, hydrostatic pressure values corresponding to the onset of cavity nucleation, according to the Ball’s model (Eq. (2)), are plotted in Fig. 9 against distance measured from the particle pole to specimen edge were increment displacement is applied. As expected, independently from the particle size, this pressure is maximal near the reinforcing particle pole. Furthermore, it quickly decreases as one moves slightly away from this pole and then tends to reach a horizontal asymptotic value. One can also note that the smaller the particle size, the more accentuated this decrease. These results confirm experimental observations reported by Oberth and Bruenner (1965), Gent and Park (1984), assuming that a critical hydrostatic pressure governs the cavity nucleation.

Although results are only shown for three volume fractions, the same trends were observed for all the studied cases.

5. Conclusion

In this work, the fracture of rubber-like materials by the cavitation phenomenon has been experimentally and numerically analysed.

Specific disc-shaped samples, called pancake specimens, were experimentally tested under uniaxial tension. Because of the particular geometry of such specimens, high stress triaxiality seems to be generated in the bulk of the material, increasing the hydrostatic stress and, therefore, leading to the cavity nucleation. In fact, fracture surfaces exhibit void footprint suggesting that the total rupture of the specimen happened by the nucleation and growth of cavities. The volume fraction and size of the observed voids depend on the specimen shape factor. In fact, increasing the shape factor of the specimen allows the occurrence of small cavities uniformly distributed through the fracture section. However, when

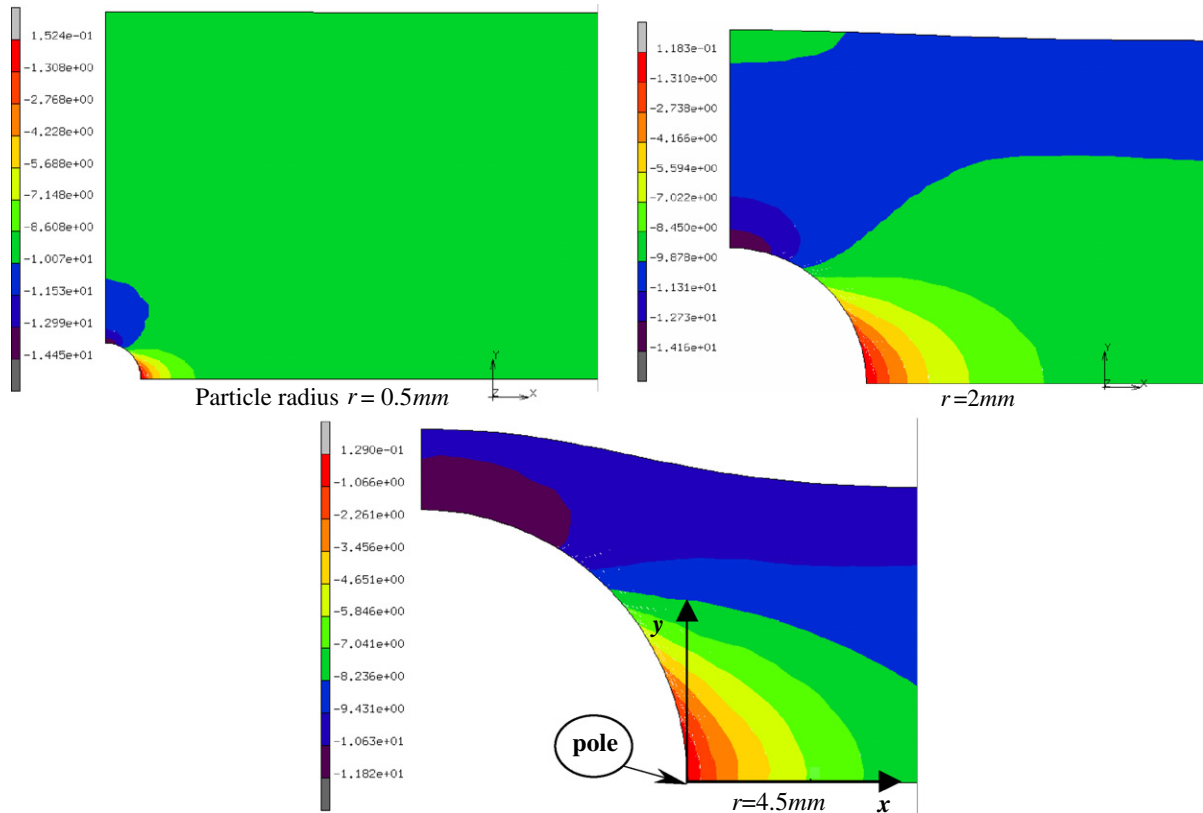


Fig. 8. Distribution of the factor $p - 5E/6$ around a rigid inclusion embedded in a rubber matrix.

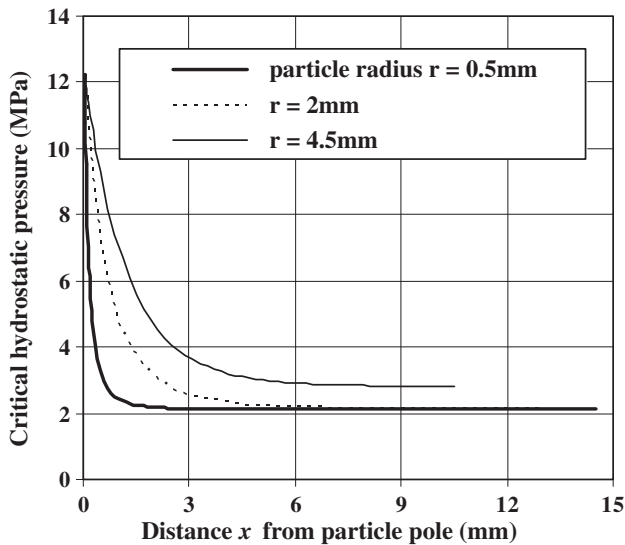


Fig. 9. Theoretical critical hydrostatic pressure against distance measured from the particle pole.

this factor decreases, the density of voids decreases while their size increases. Moreover, it has been proved, by measuring the specimen volume variation, that the apparition of a critical fraction of cavities is accompanied by a slope break of load–displacement curves. This slope break is very marked for high shape factor values.

The FE study allowed the backing up of the experimental results analysis. It also permitted to further understand the fracture of elastomers by cavitation process. In fact, it highlighted effects of

specimen shape factor and those of filler volume fraction on cavitation in such materials. In short, FE analyses mainly showed that voids initiate at the specimen central zone. The size, number and the distribution of these voids dependent on the specimen shape factor. Moreover, FE analyses proved that, in reinforced elastomers, higher triaxiality is involved at the pole of the inclusion suggesting that cavities first nucleate in this region.

Most of our results are in conformity with experimental observations reported in the literature.

In the future, improvements in the detection of the void nucleation will be very helpful. It will be also interesting to develop methodologies allowing analysis of cavitation in elastomers by taking interaction effects between fillers into account. Such methodologies could be based on homogenisation methods or on micro–macro approaches. Furthermore, the approaches based upon fracture mechanics concepts may be useful when dealing with the irreversible growth of a pre-existing cavity. So, if such a criterion is combined with a void nucleation one, it should perhaps be possible to describe as well as the void nucleation phase and its stable but irreversible growth. A void-growth constitutive model for rubber-like materials will be presented in a forthcoming paper using the theoretical methodology given in (Zairi et al., 2008, 2011) and the fruitful experimental results of this study. The strong effect of the surface tension on the void growth requires to be taken into account especially in the beginning of growth process, i.e. when the voids are small (Fond, 2001; Zairi et al., 2005). Therefore, an accurate estimation of this property is needed.

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