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Experimental real-time tracking and diffusion/mechanics numerical simulation of cavitation in gas-saturated elastomers

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ABSTRACT

The effect of a gas decompression after saturation on rubbers was investigated with both an experimental and a numerical approach. Experimental results have been obtained with a commercial transparent rubber under hydrogen and a tensile machine fitted with a pressure chamber that allows a spatial and temporal tracking of damage. The influence of the decompression rate and cavity radius have been studied and compared with a numerical model based on the theory of the hollow sphere and the implementation of a Fickean law to have gas diffusion in the material. This multi-scale model was used to temporally predict the response of a spherical cavity in a hyperelastic incompressible material submitted to a coupled gas/mechanical loading. The aim of the study was to understand the gas exchanges between the cavity and the material and to temporally predict with a simple critical stretch ratio criterion when a small cavity can growth to a visible size. The influence of decompression rate, pressure level, cavity radius and position in the sample were discussed and compared with experiments.

1. Introduction

Gas-saturated rubbers undergo very detrimental cavitation damage during fast decompression, caused by the expansion of the sorbed gas (Denecour and Gent, 1968). This decompression-induced cavitation mode is often referred to as eXplosive Decompression Failure (XDF). The phenomenon has been experimentally shown in several rubber-gas systems, among which carbon dioxide (Briscoe and Liatsis, 1992; Briscoe et al., 1994; Embury, 2004; Zakaria and Briscoe, 1990), argon (Gent, 1990; Gent and Tompkins, 1969; Stewart, 1970), methane (Stevenson and Morgan, 1995) but mechanisms responsible for damage are not understood yet. A few modeling predictions were proposed, but in very restricted and drastic conditions as detailed below in the Introduction. However, XDF is a major issue for several application fields like offshore industry, carbon dioxide storage and hydrogen energy facilities. Only few authors investigated this phenomenon in hydrogen (Yamabe and Nishimura, 2009; Yamabe and Nishimura, 2012).

When rubbers are exposed to high gas pressure, no damage appears during the saturation stage (Stewart, 1970). In the previously investigated gas-polymer systems, a few cavities appear during decompression or a few minutes later. Satellite bubbles further nucleate around the primary ones, due to a local stress concentration (Gent, 1990). When decompressed from a low saturation pressure (P_{sat}), rubber samples do not exhibit any damage. From higher saturation pressures, the density of cavities increases with P_{sat} (Stewart, 1970) except near surfaces where there exists a boundary layer without any visible damage (Stevenson and Morgan, 1995; Zakaria and Briscoe, 1990). Except for a very small number of reported works (Major et al., 2007), XDF was tracked after complete and uncontrolled decompression and removal of the sample from the pressure chamber.

Cavitation criteria developed to predict this phenomenon are mainly transferred from pure mechanical framework. Indeed, mechanical cavitation in rubber-like materials is well known since the Poker Chip Test of Gent et al. in 1959 (Gent and Lindley, 1959). The Poker Chip Test consists in inserting a thin cylinder of elastomer between two metal cylinders, which are pulled apart, creating hydrostatic stress at the center of the sample. More recently, such damage under a merely mechanical loading has been experimentally observed in real time in a transparent rubber (Cristiano, 2009). When the hydrostatic pressure was higher than 5 $\mu/2$ where μ stands for the shear modulus, cavities appeared in the center of the specimen (Gent and Lindley, 1959). This critical pressure was based on the calculation of Green and Zerna in a hollow sphere under hydrostatic loading (Green and Zerna, 1954). Other studies postulated the existence of defects in the material (Butler et al., 1998; Gent and Lindley, 1959) and modeled the sudden growth of this pre-existing defect as a hyperelastic hollow sphere problem (Ball, 1982). More sophisticated models were published recently to take into account shear stresses effect and predict onset-of-cavitation surfaces upon

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three-dimensional loading (Lopez-Pamies et al., 2011). The influence of the rubber constitutive law has been addressed, considering a compressible material (Murphy and Biwa, 1997; Shang and Cheng, 2001), an incompressible material (Dollhofer et al., 2004; Hou and Abeyaratne, 1992; Jin et al., 1999) or a viscous material (Lee and Mear, 1994). Some of these works took into account the surface tension (Dollhofer et al., 2004; Gent and Tompkins, 1969; Huo et al., 1999). In most studies, the problem was solved for hydrostatic loadings only, in a uni- or bi-axial framework. Only a few works generalized the problem to tri-axial loadings (Diani, 2001). The aim of these approaches was to predict a critical mechanical loading for cavitation onset. None of them were able to predict time effects, such as the loading rate sensitivity. Previously reported models were not able to predict damage of elastomers under a real gas cycle including partial decompression or to account for the influence of the thickness of seals submitted to coupled diffusion/mechanical loading.

Since rubbers are used in many industrial applications under various loadings, it is crucial to predict the XDF sensitivity to the loading history among which saturation pressure and decompression rate. In the case of XDF, time effects are important. For instance, they may arise from variable decompression rates. In a methane-elastomer system, Stevenson and Morgan (1995) evidenced that the density of cavities does not depend on decompression rates between 40 MPa/min and 69 MPa/min, whereas it decreases by a factor of 5 between 40 MPa/min and $2 \cdot 10^{-3}$ MPa/min. It was interpreted as a gas diffusion effect. In general, there are only a few works aiming at predicting cavitation onset in a coupled diffusion/mechanics framework. Yamabe and Nishimura (2009), Yamabe and Nishimura (2012) assumed a pre-existing cavity that mimicked clusters of hydrogen molecules formed at the very early stage of decompression. They also calculated the critical internal pressure to enhance fracture of the cavity wall. They considered that no desorption occurred so that they solved a time-independent problem. In the 70's, Stewart (1970) accounted for the gas diffusion at the surface of a similar cavity, proposed an analytical solution that gives the time history of a cavity submitted to a fast decompression as a function of the cavitation time. Assuming a cavitation time and a gas concentration profile around the cavities, this solution was able to reproduce the qualitative dependence on gas diffusion parameters and saturation pressure.

In this context, the present study focuses on the very early stage of damage and has two objectives. The first one is to complete the experimental characterization of the phenomenon in extended conditions, i.e. by real-time tracking *during* the decompression stage, and to map the influence of key loading parameters like saturation pressure and decompression rate on the cavitation onset. A key contribution of the present work is to access the current pressure at cavitation onset and thus to discuss the relevance of hydrostatic pressure-based cavitation criteria. The second one is to improve the prediction of cavitation in the same extended conditions. In particular, a major originality is to compute the volume change of a pre-existing spherical cavity located in the core of the sample during decompression, in order to temporally predict XDF. Gas diffusion and mechanics are both solved in a hollow sphere problem with boundary conditions calculated from the macroscopic coupled diffusion/mechanics problem at the scale of the sample. This radially symmetric framework is relevant since only pure hydrostatic mechanical loadings are considered. In addition, our numerical model can take into account the distance from the cavity to the free surface of the sample, which has never been modeled before.

The present study is based on vinyltrimetoxysilane interacting with hydrogen but the approach and results more widely concern the whole field.

2. Experimental

2.1. Material and gas

Decompression experiments were conducted in a transparent and un-filled vinyltrimetoxysilane previously saturated by gaseous hydrogen. The solubility s_p and diffusion D coefficients of hydrogen into this rubber were measured using the Thermal Desorption Analysis (TDA) technique of Yamabe and Nishimura (2009) in cylindrical samples (height $z_0 = 2$ mm; radius $r_0 = 6.5$ mm). The hydrogen desorption kinetics was analytically fitted from the series proposed by Demarez et al. (1954):

$$Qu_{H2}^{RenT} = \frac{32}{\pi^2} V s_p \left[\sum_{n=0}^{\infty} \frac{exp(-(2n+1)^2 \pi^2 Dt/z_0^2)}{(2n+1)^2} \right] \times \left[\sum_{n=1}^{\infty} \frac{exp(-D\beta_n^2 t/r_0^2)}{\beta_n^2} \right] \quad (1)$$

where Qu_{H2}^{RenT} stands for the hydrogen quantity remaining in the volume V of the sample, βn is the root of the $J_0(\beta) = 0$ equation with J_0 Bessel function. Coefficients D and s_P were fitted from a classical genetic algorithm (Renner and Ekart, 2003).

The optimized values were $4.22 \cdot 10^{-2}$ cm^3(STP) cm^{-3} bar for the solubility s_P and $2.4 \cdot 10^{-9}$ m^2 s^{-1} for the diffusion coefficient D. STP stands for Standard conditions of Temperature (273 K) and Pressure (0.1013 MPa). These fitted values were in good agreements with data reported about hydrogen-rubber systems (Hagg, 2000).

The hyperelastic response of the rubber was calibrated from tensile Dynamic mechanical analysis (DMA) and monotonic tension tests. A shear modulus of 0.4 MPa was measured at room temperature with both tests. It could be argued that these mechanical tests were performed in air and further applied to the hydrogen-saturated rubber, but hydrogen was shown to have a negligible effect on the mechanical properties of rubber materials (Yamabe and Nishimura, 2011). No other transition than the glass transition –occurring at −68 °C– could be detected from the DMA analysis, and no damping contribution was measured at room temperature. Optical microscopy did not evidence any pre-existing micro-cavity. However, sub-micron cavities or defects smaller than 1 μm could not be detected from such optical techniques.

2.2. Experimental results

Decompression tests were carried out in an Instron 8802 tensile machine fitted with a pressure and temperature regulated chamber which allows mechanical testing in gaseous nitrogen, hydrogen or carbon dioxide. For details about the experiment, one can refer to previous papers from the same laboratory (Castagnet et al., 2012). Regarding the state-of-art, a key contribution of the present work dealt with the ability to simultaneous spatial and temporal tracking of XDF. It was made possible by two sapphire windows inserted in the front and back doors of the chamber.

Decompression tests consisted of a pressurization stage at constant pressure rate, followed by a constant pressure stage aimed at stabilizing the temperature (at about 20 °C) and saturating the sample. Finally, the decompression step was conducted under constant pressure rate. All tests were performed at room temperature, excepted that temperature variations resulting from the pressure history were not regulated. Experimental conditions, e.g. saturation pressure, pressurization and decompression rates, did not enhance any phase transformation of the hydrogen gas.

Two main series of experiments were carried out. In the first one, the saturation pressure varied from 3 up to 27 MPa and decompression always occurred at 9 MPa min^{-1}, whereas in the second series, samples were decompressed at a constant rate varying from 0.1 up to 90 MPa min^{-1} from an always identical saturation pressure of 9 MPa. In this way, the influence of saturation