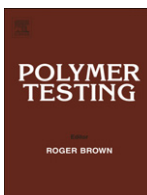




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## Material behaviour

# Structural characteristics and swelling mechanism of two commercial nitrile-butadiene elastomers in various fluids

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### ABSTRACT

Results on the structural characteristics and swelling mechanisms of two commercial elastomers, viz. N0674-70 and peroxide cured N0741-75, in a variety of fluids (toluene, o-xylene, p-xylene, mesitylene, HISOL-15, n-hexane, ethanol, tert-butyl alcohol, acetone, JP-8, S-8 and blends of S-8 plus HISOL-15 and S-8 plus toluene) are reported. Both changes in mass and volume of rectangular test pieces were monitored as a function of time. Structural characterization of the elastomers was done using x-ray diffraction and FTIR spectroscopy. Time dependence of the recovery of swelling after the elastomers were taken out of the fluids was monitored by x-ray diffraction and changes in mass. In general, swelling of the peroxide cured N0741-75 was less than that of N0674-70, percentage changes in mass were less than those in volume, and the swelling is strongly fluid dependent. For initial times, the swelling is interpreted in terms of diffusion of the fluid into the elastomer. However, diffusion is not strictly Fickian since the exponent, varying between 0.4 and 0.6 for different fluids, differs from 0.5 expected for Fickian diffusion. It is shown that the results of maximum swelling can be interpreted in terms of compatibility of the Hansen solubility parameters  $\delta_T$  of the fluid and the elastomer, swelling decreasing on a Gaussian curve as  $\delta_T$  of the fluid deviates from that of the elastomer. Also, swelling is determined by the polybutadiene component of the elastomer. In HISOL-15, a commercial aromatic solvent, nitriles swell by about 55%, and mixing of HISOL-15 with S-8 produces swelling linearly proportional to the % of HISOL-15. The observed few percent decrease in swelling on long-term exposure may be related to loss of material from the elastomers.

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## 1. Introduction

The engines in most modern transportation vehicles used in the civilian and military sectors are designed for fuels derived from conventional petroleum crude oil. These fuels contain a certain percentage of aromatics and sulfur that provide lubricity and the desired amount of elastomer swelling of the O-rings to prevent fuel leaks. The synthetic fuels produced by Fischer-Tropsch synthesis, if used in such engines, will have to be blended with appropriate additives to achieve the desired amount of lubricity and elastomer

swelling. Therefore, it is highly desirable to investigate and understand the swelling characteristics of commercial elastomers in different fluids.

Nitrile rubber elastomers are copolymers of polyacrylonitrile,  $-(CH_2=CH-C\equiv N)_n$  and polybutadiene,  $-(CH_2-CH=CH-CH_2)_n$ . The characteristics of a given elastomer are, therefore, expected to depend on the percentages of these two primary components in the elastomer. In recent years, results from a number of investigations have appeared in literature [1–5] in which the authors have varied the relative compositions of these two components in laboratory prepared samples and investigated their swelling characteristics in aromatics such as benzene, toluene and xylene. These studies have

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provided considerable insight into factors such as the role of molar volume of the fluid on its diffusion into specimens of various compositions [1–5].

Commercial elastomers (e.g. O-rings) usually contain additional components such as plasticizers, fillers and curing agents in addition to polyacrylonitrile and polybutadiene. In addition, the relative percentages of these components in a given elastomer are usually a trade secret and not known accurately. A few years ago, Muzzell et al. [6] carried out investigations of the swelling and compatibility of commercial elastomers in synthetic S-5 fuel blended with various percentages of the commercial A-150 aromatic solvent and in JP-5/JP-8 aviation fuels. In these investigations, magnitudes of the saturation swelling achieved after several days of immersion in a fluid was the primary focus of attention. This work was followed up by investigations by Graham et al. [7] on the swelling characteristics of commercial nitrile O-rings in selected aromatics blended in synthetic jet fuel.

The present study was undertaken to investigate the swelling characteristics of the same two elastomers as used in the work by Muzzell et al. [6] with the aim of reaching a more fundamental understanding of how saturation is achieved in different fluids and how it depends on the characteristics of the fluids. For this purpose, we have used a variety of fluids, both aromatics and alkanes, and measured the time dependence of changes in mass and volume of the elastomers. The results are discussed in terms of the diffusion phenomenon and on the properties of the fluids such as their molar volumes and their Hansen solubility parameters (HSPs) [8]. Results on the swelling characteristics of the elastomers in thirteen different fluids and fluid blends are reported. The compatibility of the HSPs of an elastomer and those of a fluid are shown here to control the observed swelling.

## 2. Experimental procedures

### 2.1. Materials

Two commercially available elastomers, viz. N0674-70 and peroxide cured N0741-75 from Parker Co., were studied. These samples are identical to those used by Muzzell et al. [6] in their investigations of the saturated swelling of these elastomers. These two nitrile elastomers contain polyacrylonitrile and polybutadiene in various proportions in addition to fillers to provide strength, and they are typically found in some fuel systems of army tactical and combat vehicles [6]. The solvents used for the experiments were toluene, o-xylene, p-xylene, mesitylene, HISOL-15, n-hexane, ethanol, tert-butyl alcohol, acetone, JP-8, S-8 and blends of S-8 plus HISOL-15 and S-8 plus toluene. HISOL-15, a universal aromatic solvent, was kindly provided by SHELL Co. Sheets of the two elastomers and the fluids JP-8 and S-8 were kindly provided by Pat Muzzell. All other fluids were purchased from commercial sources (Alfa Aesar).

### 2.2. Procedures for measuring swelling

Small rectangular test pieces were cut from the available elastomer sheets by means of a sharp knife. The thickness,

length and width of the test pieces were measured using a high-precision dial-gauge caliper with an accuracy of  $\pm 0.01$  mm with typical dimensions of samples being about  $10 \times 10 \times 2$  mm<sup>3</sup>. All measurements were done at room temperature. Since the measured changes in the dimensions of the test pieces during swelling in most fluids are much larger than the experimental precision of  $\pm 0.01$  mm, this simple method provides quite reliable data. For mass changes, we followed the ASTM D471 (ISO 1817) method using a sensitive balance with accuracy of  $\pm 0.1$  mg. The test pieces were immersed in about 25 ml of the solvent in small glass test bottles. To measure changes with time, the immersed samples were periodically removed from the glass bottle and excess solvent wiped off quickly using wipes. The sample was then weighed and its dimensions measured, followed by immediately placing the sample back in the fluid. The time taken for each set of measurements was kept to a minimum and was approximately 60 s.

## 3. Structural characterization of elastomers

### 3.1. Fourier Transform Infra-Red Spectroscopy (FTIR)

A Mattson Infinity Gold FTIR spectrometer was used to determine the IR spectra of the two elastomers and samples of pure polybutadiene and raw polyacrylonitrile butadiene to examine the differences in the chemical structure of the commercially available elastomers vis-a-vis the pure components. Techniques of attenuated total reflectance (ATR), photoacoustic spectroscopy (PAS) and transmission spectroscopy using the standard KBr method were tried to obtain the best signal to noise (S/N) in the spectra. For PAS and KBr methods, the samples were powdered by hand grinding onto white paper to obtain very fine powder of these difficult-to-grind polymers.

The IR spectra of polybutadiene and raw polyacrylonitrile-butadiene obtained by using ATR are shown in Fig. 1. A prominent feature of polyacrylonitrile butadiene is the presence of the nitrile (cyano)  $\text{C}\equiv\text{N}$  group which is known to yield a sharp peak at  $2235\text{ cm}^{-1}$  in

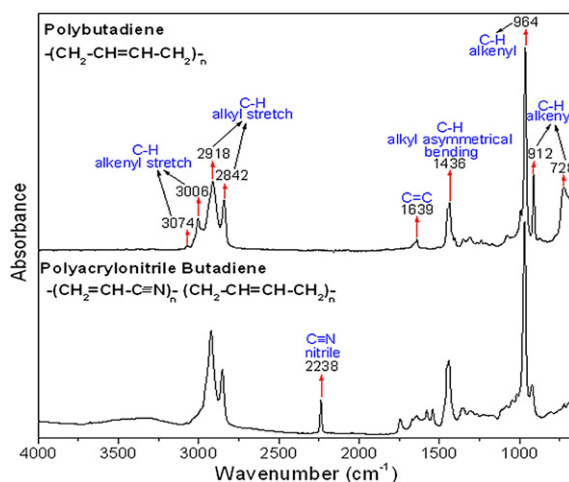


Fig. 1. IR spectra of polybutadiene and raw polyacrylonitrile butadiene acquired using ATR.

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