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The Effect Hydrogen has on Polymers Used as Seals and Liners in the Hydrogen Infrastructure

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E = EPDM N = NBR
F P = Filler and plasticizer
nF P = No filler with plasticizer
F nP = Filler without plasticizer
nF nP = No filler and no plasticizer

Introduction

- Polymeric materials are used for gas seals and liners in hydrogen equipment [1].
- This study focuses on how ethylene propylene diene monomer (EPDM) and nitrile butadiene rubber (NBR) are affected by pressurized hydrogen at different formulations of filler (carbon black and fumed silica, both too small to image) and plasticizer.
- X-ray computed tomography (XCT) was used to non-destructively reconstruct 2D images of an object into a 3D data set which can be rendered into still images or videos [2].
- Four formulations were tested for each polymer type, resulting in eight total samples in addition to a blank for each sample. The formulations tested are summarized in the key above.
- Videos of XCT images show voids and ZnO particles for each sample. Some samples showed cracks and agglomerations of fumed silica. No blank samples had either voids or cracks.
- Cumulative void volume distribution was calculated for 2 of the 8 hydrogen-exposed samples. Total void fraction, for all 8.

Methods

- XCT images were taken on a Zeiss Xradia 520 Versa.
- Each exposed sample was subjected to 15 kpsi (100 Mpa; 1000 atm.) hydrogen for 7 days at room temperature.
- Wondershare's Filmora video editing software was used to view, edit, and display videos. Scan the QR code to the right for examples of the videos.
- In Avizo 9.5, the data sets were segmented into four regions; Exterior, Voids, Matrix, and ZnO by radiograph intensity, which varied for each sample requiring custom parameters.
- Volumetric statistics for the voids were then extracted allowing comparison of void fractions, see Figures 2 and 3.

Results

- E F P has the least total percent void volume at 0.12%.
- N nF P has the lowest percent void volume at 0.15%.
- 63% of the cumulative void volume in E nF nP has a larger equivalent diameter than the largest void found in E F P (Figure 3).
- The largest void in E nF nP is more than 6 times larger than the largest void volume in E F P (derived from Figure 3)
- Inspection of the XCT images indicates that E nF nP has developed cracks not seen in E F P but not as extensive as in E nF P (Figure 1).

Data

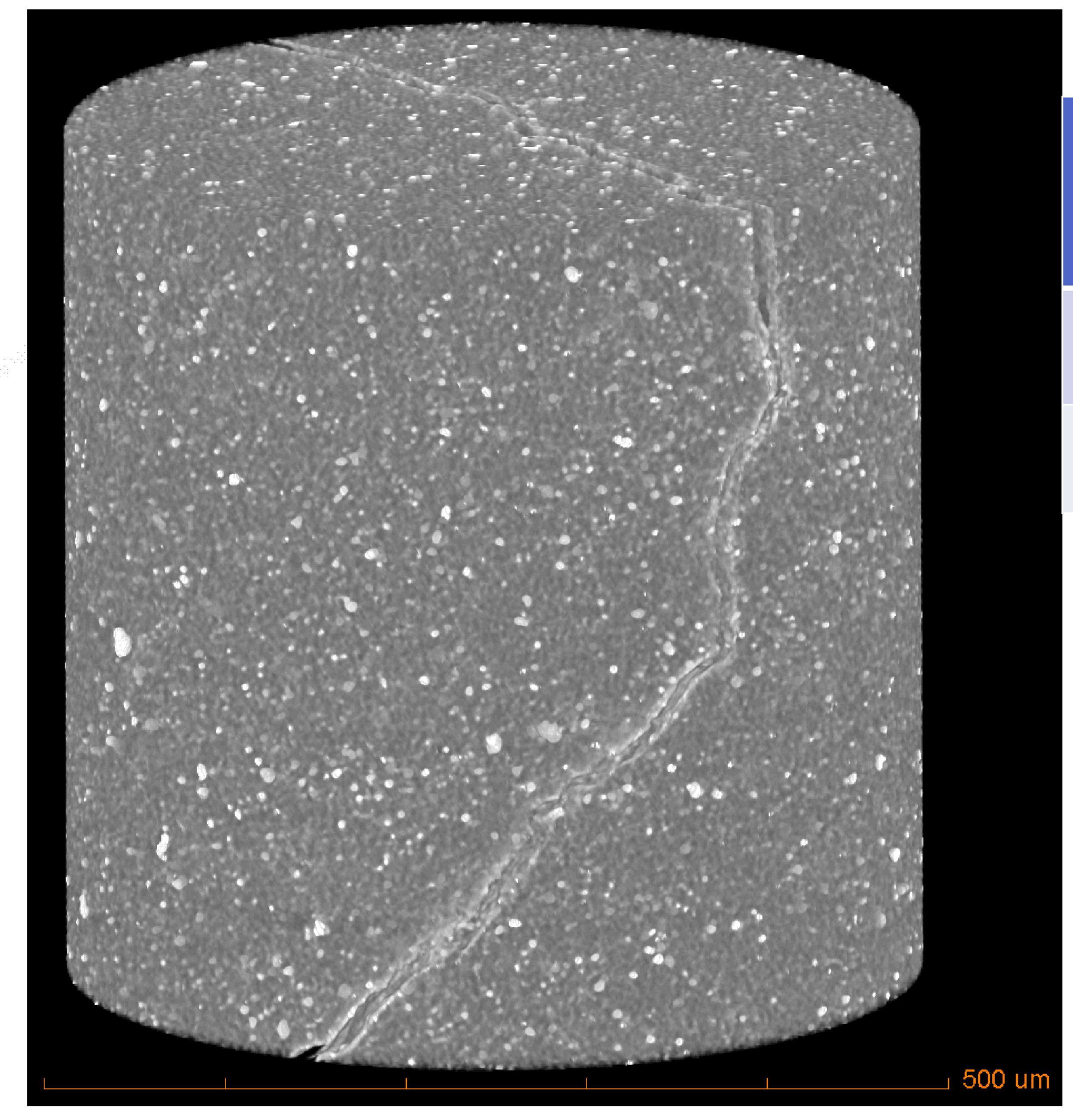

Figure 1: An XCT image of an interior cylinder about 0.5 mm in diameter of E nF P, showing the only example of extensive cracking in this experiment. This sample also has the largest void volume.

Sample	D10 (µm)	D50 (µm)	D90 (µm)
E F P	1.33	6.71	17.4
E nF nP	1.98	25.4	37.1

Table 1: The equivalent spherical void diameter at 10%, 50%, and 90% cumulative void volume as shown in Figure 3.

Use the QR code above to access a series of movies of the hydrogen-exposed samples.

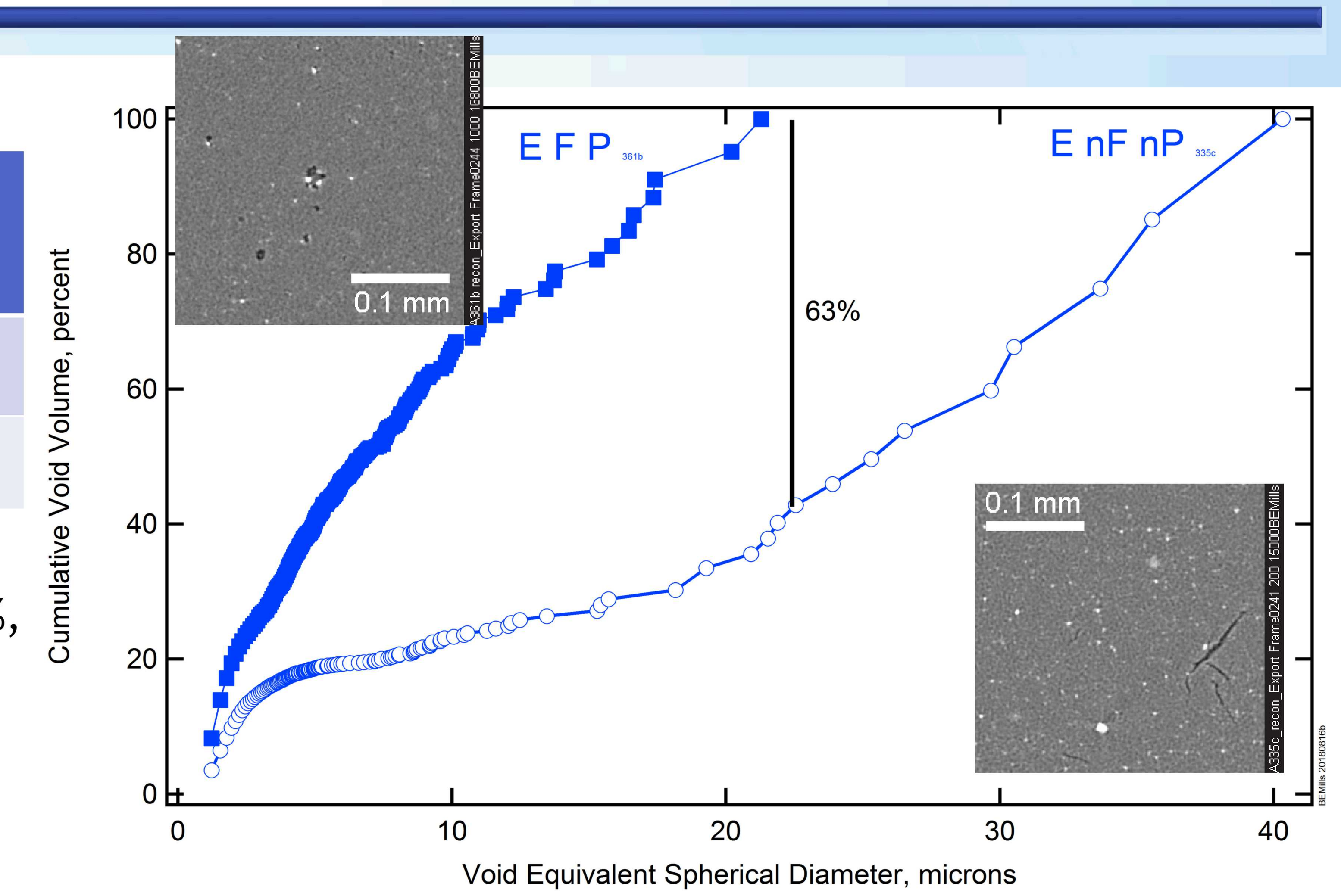

Figure 3: Cumulative void volume distributions for E F P and E nF nP. There are more larger sized voids in E nF nP than there are in E F P, showing the advantage of filler and plasticizer. The largest volume voids are cracks, hence the use of void equivalent spherical diameter.

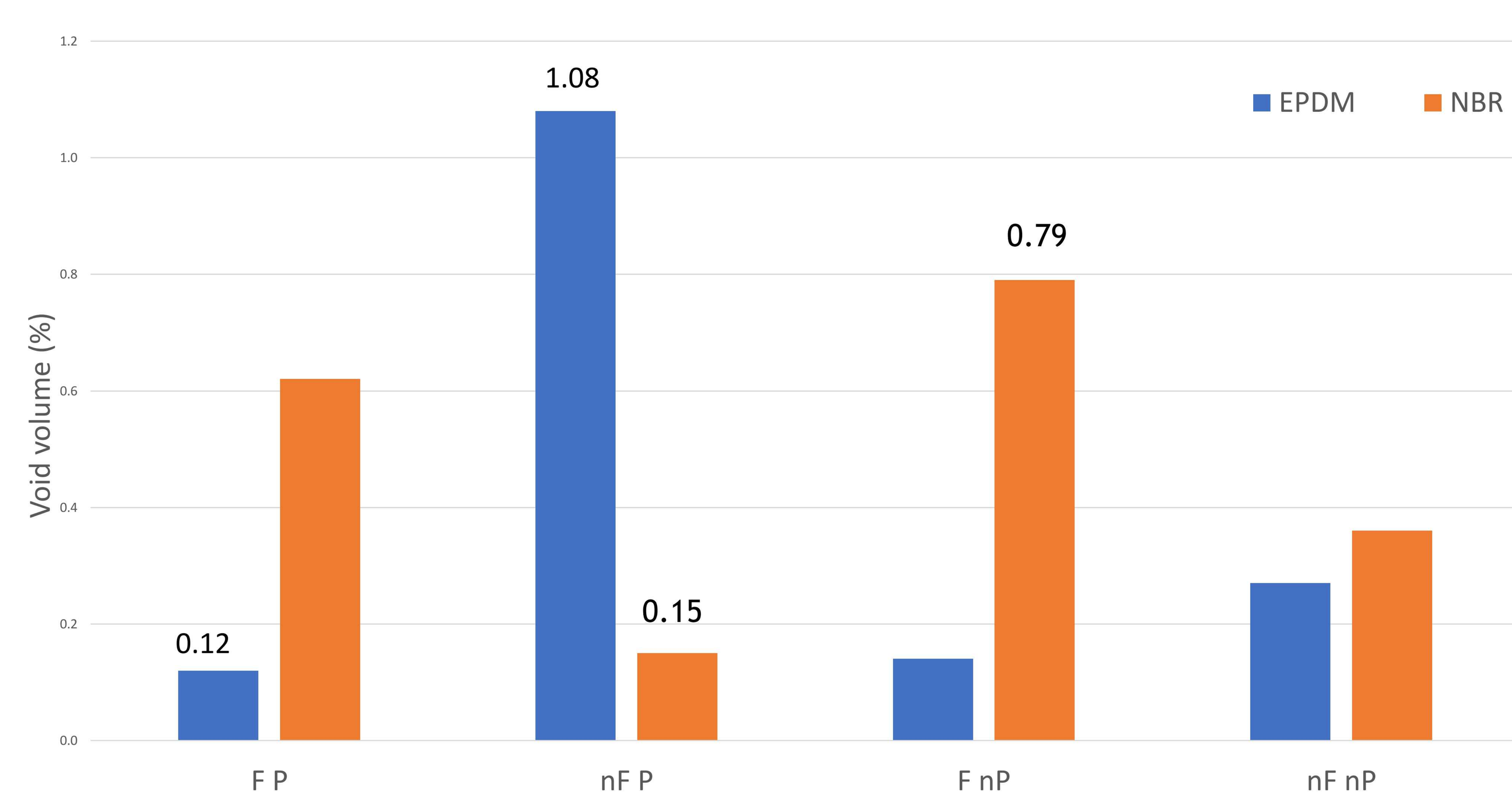

Figure 2: Percentage void volume of like formulations of EPDM and NBR polymers. The highest and lowest percentage for each polymer are indicated on the graph. Note that without filler EPDM cracks badly (see Figure 1).

Discussion

The ultimate goal of this research is to generate a fundamental understanding of the effect fillers and plasticizers have in how polymers, specifically elastomers, are affected by high pressure hydrogen. Here, we have developed a method to determine void fraction for eight model material formulations and the void size distribution for two of these formulations.

- The highest void volume is 1.08% in E nF P due to the extensive cracking as seen in Figure 1.
- E F P has fewer large sized voids than E nF nP (Figure 3).
- Plasticizer alone is advantageous for NBR but deleterious to EPDM.
- Filler alone is advantageous to EPDM but deleterious to NBR.
- EPDM benefits from both filler and plasticizer, while NBR does not.

Possible future work based on these data:

I. Determine the cumulative volume percent distribution for the other six samples, as seen in Figure 3 for two examples.
II. Locate where in the sample voids are most likely to form (e.g. near an original or cut surface or in the interior).
III. Determine what fraction of voids form around ZnO particles.
IV. Measure the sphericity of each void to document the transition from spherical void to crack.

Acknowledgements

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Resources

[1] Mills, Bernice & Menon, Nalini. "High Pressure Hydrogen Testing of Materials for Seals and Liners". Sandia National Laboratories.
[2] Clayton, Carmen, Mills, Bernice, Buffleben, George, & Vu-Nguyen, Thien. "Enhanced Preesentation of Tomographic Data". Sandia National Laboratories.