

# A Study on Sealing Behavior of Rubber O-Ring in High Pressure Hydrogen Gas

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## ABSTRACT

Rubber O-rings installed in hydrogen tanks for fuel cell electric vehicles are repeatedly exposed to high pressure hydrogen gas. Exposure to high pressure gas sometimes causes cracks as a result of blistering after decompression. The degree of blister damage is influenced by material, environmental conditions such as decompression rate, and sealing shape such as squeeze ratio. Focusing on environmental conditions out of these influential factors, in this study, a high pressure hydrogen durability tester which exposes rubber O-rings repeatedly to high pressure hydrogen gas at arbitrary test conditions was developed. Using this tester, the influence of hydrogen pressure and temperature on blister damage and permeability was investigated for sealing materials used conventionally for high pressure equipment.

## INTRODUCTION

Rubber seals, which are installed in high pressure hydrogen tanks, pipe lines and valves of fuel cell electric vehicles (FCEV), are repeatedly exposed to high pressured hydrogen gas. The pressure of hydrogen gas is very high (70MPa) and the range of ambient temperature is very wide (-70 to 100 °C); therefore, the environmental conditions for sealing are severe. The exposure of rubber O-rings to high pressured gas sometimes causes mechanical damages such as large

deformation, swelling and blistering after decompression [1-8], although there are only a few reports on blister fracture caused by high pressured hydrogen gas [9,10]. To maintain sealing performance and durability of the O-ring under the high pressured hydrogen gas, several types of gas leakages must be considered.

Figure 1 shows a schematic representation of conventional gas leakages, which can be divided into three types. One is gap leakage, which occurs at the boundary between the O-ring and equipment. Another is permeation leakage, which permeates through the O-ring. The other one is mechanical damage leakage which is caused by breaking of the O-ring. Mechanical damages can suddenly cause heavier leakage than the others. At low ambient pressures, gas and permeation leakages have significant effects on the sealing function of O-rings. At high ambient pressure, mechanical damage leakage is the most significant factor.

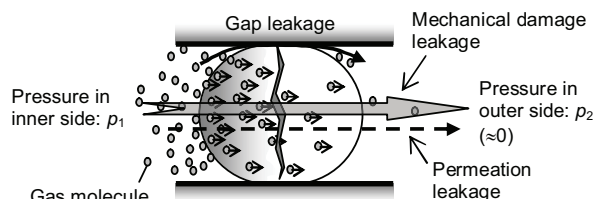


Figure 1: Schematic representations of common gas leakage types.

In general, a blister is defined as a crack or bubble initiating from the inside of a rubber material after decompression [1]. Figure 2 shows a schematic representation of cracks caused after decompression [5–8]. When a rubber exposed to high pressured gas is not squeezed, randomly distributed cracks and ring-like cracks are formed. In previous studies [9, 10], cylindrical specimens with a diameter of 29 mm and thickness of 12.5 were exposed to hydrogen gas at  $\leq 10$  MPa after which randomly distributed cracks could be seen after decompression. Figure 3 shows optical microscope images of inner cracks after decompression of transparent EPDM composite exposed to hydrogen gas [10]. The transparent specimen was exposed to hydrogen gas at 10 MPa and 30 °C for 65 hours in Fig. 3 (a), and considerable inner cracks were observed at 5 minutes after decompression. Figure 3 (b) shows the high magnification images of the transparent EPDM composite exposed to hydrogen gas at 3 MPa and 30 °C for 65 hours. While two large-sized cracks can be seen at 20 minutes after decompression, micrometer-sized

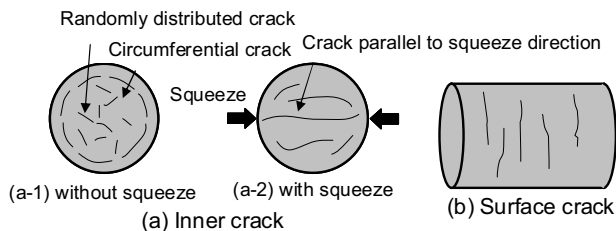


Figure 2: Cracks generated by decompression reported in early studies [5–8].

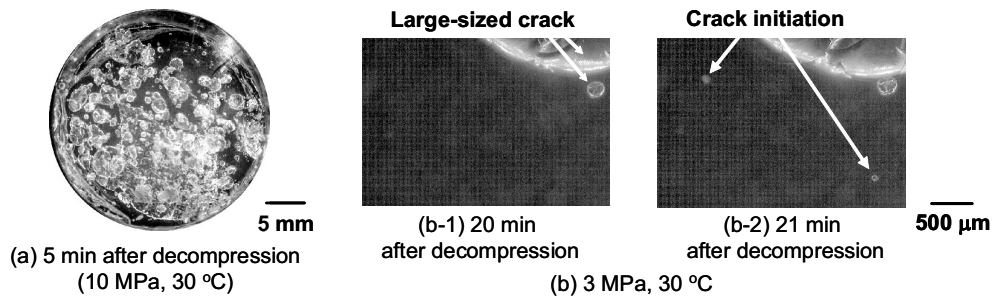


Figure 3: Optical microscope images of transparent EPDM composite [10].

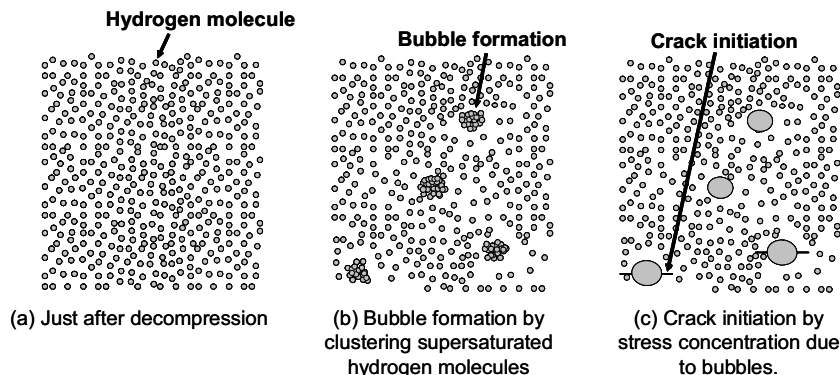


Figure 4: Model of crack initiation [10].

cracks formed at 21 minutes in addition to these large-sized cracks. Figure 4 shows the model of crack initiation. The cracks shown in Figs. 3 (a) and (b) are thought to have initiated due to stress concentration of micrometer-sized bubbles formed by supersaturated hydrogen molecules in the composite after decompression [10]. As for the ring-like cracks, it has been reported that shear stress due to hydrogen concentration gradient at decompression influences the generation of this type of cracks [7]. When a rubber material is squeezed, cracks parallel to squeezing are formed due to tensile stress generated at the center of a specimen. Moreover, when decompression rates are rapid, more than 1 MPa/sec, surface cracks may also be formed.

Figure 5 shows the relationship between true fracture strength, hydrogen content, and degree of blister damage of EPDM composites in previous studies [9, 10]. Silica and carbon black were employed as fillers. The true fracture strength of these composites was calculated from nominal fracture strength and fracture stretch under incompressibility. The hydrogen contents were the values of the composites exposed to hydrogen gas at 10 MPa and 30 °C, which were measured by gas-chromatograph mass spectroscopy. No blisters were observed in the EPDM composite filled with silica due to higher true fracture strength and lower hydrogen content, while several blisters occurred in the EPDM composite filled with carbon black due to higher hydrogen content despite higher fracture strength. Material strength and hydrogen penetration properties such as hydrogen content influence blistering of rubber materials.

As known from the above result, although material properties such as material strength and hydrogen content influence the degree of blistering, the blister damage of an actual rubber O-ring is influenced by environmental condition and sealing shape as well. For example, with increasing decompression rate, the damage of the blister generally tends to be more serious. Figure 6 shows influencing factors on blistering. It can be seen that many factors influence blister damage.

From this viewpoint, this study focused on environmental conditions. In order to evaluate the influence of environmental conditions on blistering, a high pressured hydrogen durability tester for rubber O-rings which enables repeated exposure of the O-rings to high pressure gas (Maximum pressure: 90 MPa) and temperatures ranging from -60 to 100 °C was developed. Using this tester, cyclic tests at several hydrogen pressures under specific ambient temperatures were conducted and the influence of hydrogen pressure and temperature on blister damage and permeability was investigated.

## EXPERIMENTAL EQUIPMENT

Figure 7(a) shows a schematic representation of the high pressured hydrogen durability tester developed in this study. Hydrogen gas at 5 MPa is compressed to 95 MPa by a compressor and the pressurized hydrogen gas is accumulated in the pressure accumulator. After this, the high pressure hydrogen gas is repeatedly supplied to 8 O-ring specimen holders installed with O-ring specimens, and pressure cycle tests are conducted. The tester is equipped with a hydrogen concentration sensor, and the amount of hydrogen gas penetrating the O-ring specimens during the cycle test is monitored. This tester can be operated at hydrogen pressures ranging from 6 to 90 MPa, and temperatures ranging from -60 to 100

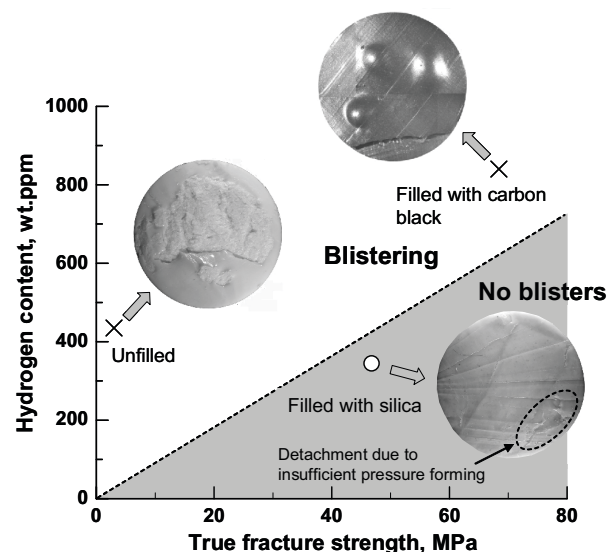


Figure 5: Influence of hydrogen content and fracture strength on blistering of EPDM composites<sup>[9]</sup>.

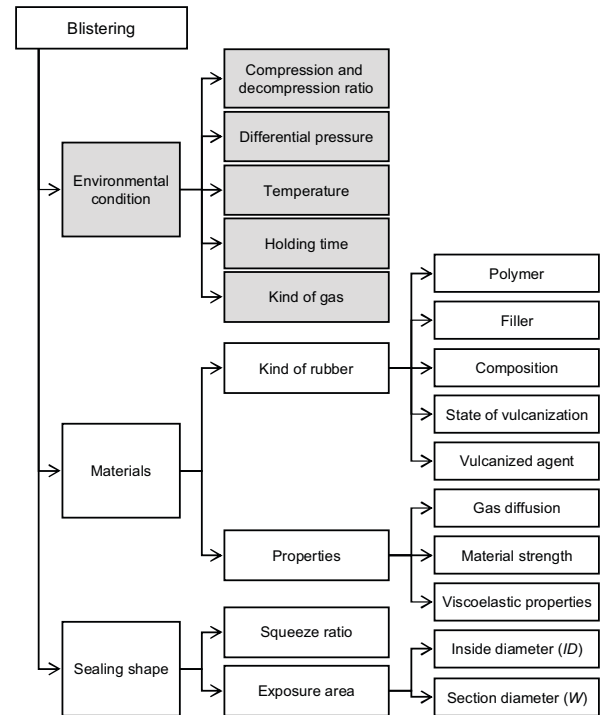


Figure 6: Influencing factors on blistering.

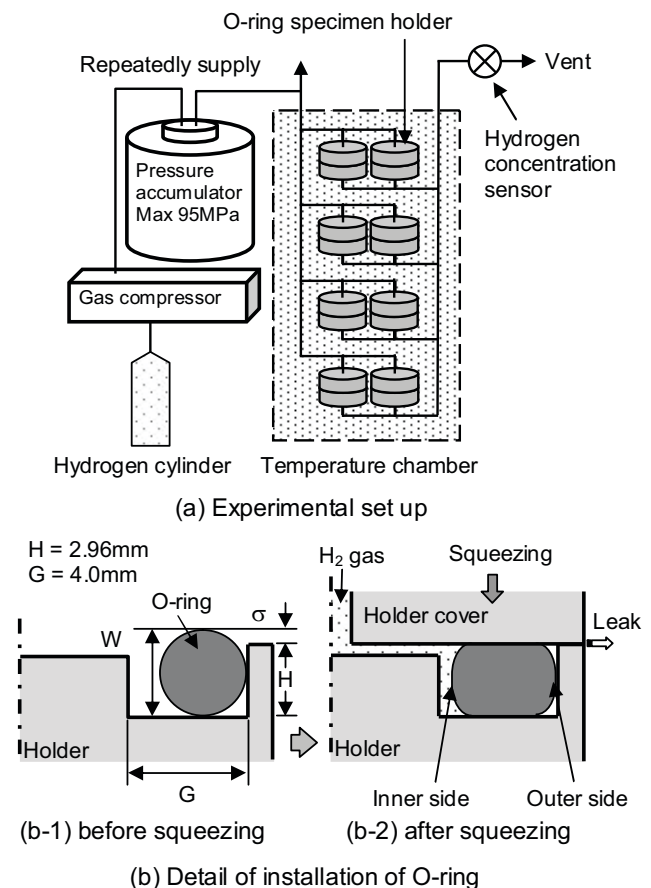


Figure 7: Schematic representation of high pressure durability tester.