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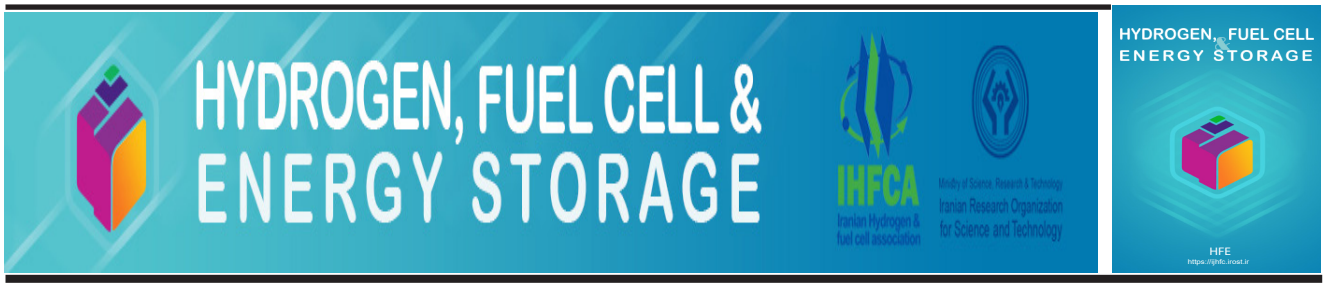

Mechanical and Chemical Characteristics of PEM Fuel Cell Sealants in a Simulated Environment

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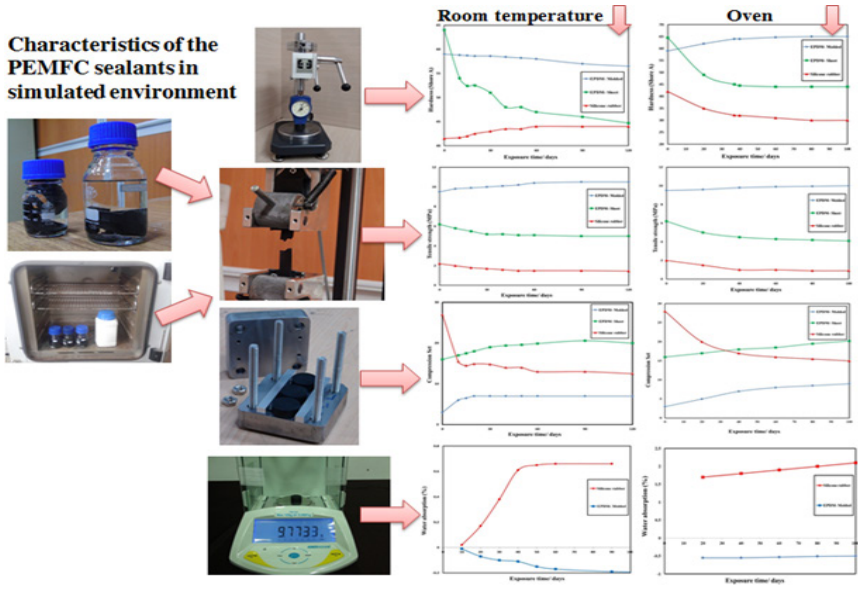

Abstract

Sealants are one of the most important components of proton exchange membrane fuel cells (PEMFCs). They play significant roles in fuel cells' safety, energy density, durability, and performance. Thus choosing the proper kind of sealant suitable for PEMFCs is essential for the performance of the fuel cells.. The seal's durability gives it the ability to correctly perform the sealing function in the fuel cell environment for an extended amount of time with only small changes in its physical and chemical properties. In this paper, the mechanical properties of three materials frequently used for fuel cell sealing are assessed in an environment similar to actual fuel cells. These three materials are silicone, EPDM (ethylene propylene dyne monomer) sheet, and molded EPDM. Mechanical properties of the materials were obtained after being used in an environment resembling a fuel cell for a specific time and temperature. The specimens' mechanical and chemical properties, such as hardness, weight changes, tensile strengths, compression set, and spectrometry, were determined in an accelerated durability test in the simulated PEMFC environment. These tests were conducted over a period of 100 days. The results revealed that molded EPDM is the best of the tested sealants based on the obtained properties in fuel cell working conditions.

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1. Introduction

Energy supply is one of the most important factors influencing a country's development. The reduction of fossil fuel resources and the pollution caused by their use have prompted mankind to search for a suitable energy production alternative [1]. Polymer fuel cells have received more attention than other fuel cells because of advantages such as low operating temperature, high power density, and low start-up time. One of the most important challenges in designing and manufacturing a fuel cell is its sealing technology [2, 3]. The PEMFCs' hydrogen (very active and explosive), oxygen, and cooling fluid make the proper sealing of fuel cell components imperative. Any leakage of these components could cause an explosion and fire, in addition to significantly reducing efficiency. The sealants must be suitably compressed to do the sealing function properly. The membrane, the gas diffusion layer, and the bipolar plates must also be sufficiently compressed to each other in order to achieve proper contact and low electrical contact resistance. If low pressure is applied on the surface of the membrane, the contact resistance increases and as a result, the efficiency of the PEMFCs decreases. On the other hand, higher pressure causes the gas diffusion layer (GDL) to be compressed excessively, which will lead to decrease gas penetration. Sealants must be chemically and physically stable perform well over the defined lifetime of the fuel cell. If any seals degrade or fail during operation or standby, reactive gases (H_2 and O_2) can leak out or mix directly. If acute, this phenomenon causes the fuel cell failure or at a lower levels causes a severe drop in the performance of the fuel cell [4, 5]. Since the long-term use of the fuel cell depends on the seal's durability, the seal material's resistance against destruction is a practical requirement to improve the life of the fuel cell. Considering the seal's importance in the fuel cell, its material should have suitable durability in the environment of the fuel cell. The seal's durability allows it to correctly perform the sealing function in the fuel cell environment for a long time with only minor physical and chemical properties changes. Different companies have introduced various materials as suitable for fuel cell seals. However, many of these materials do not perform well in real working conditions and cause problems. Using reactive materials that quickly deteriorate in the fuel cell environment wastes energy and reduces efficiency, and can cause system failure or even explode. Therefore, choosing a suitable material for fuel cell sealing is very important. Rubbers and elastomers are always the best option for the manufacturing of seals due to their structure and constituent chain. This material seals the component by being compressed. The sealing quality of elastomers is related to their chain type and the degree of reactivity of the constituent material with the environment in which they are placed [6].

Many elastomeric materials are used for sealing applications related to PEM fuel cells [7-9]. Fluoro-elastomers, silicone rubbers, and hydrocarbon-based elastomers (such as EPDM) are considered common and suitable choices for sealing compounds in fuel cells due to their excellent characteristics in terms of chemical and thermal resistance; however, each of materials has its problems. For example, fluoroelastomers show excellent chemical resistance against water and acids and also have very low hardness and compression sets, so they are expected to be very suitable and durable seals in PEM fuel cells. However, manufacturers usually do not prioritize fluoroelastomers due to their poor melt processability in injection molding and poor flexibility at low temperatures (the glass transition temperature of FKM is about -20°C). In addition, their price is also much higher than common rubbers for general use [10-12]. Silicone rubbers have good thermal resistance, but their high compression set, weak chemical resistance in acidic environments, and relatively high price compared to commonly used general rubbers have limited the use of such rubbers [13-

15]. EPDM is an important polymer with good resistance to heat, light, ozone, and UV rays and is widely used in outdoor applications. Compared to fluoroelastomers, EPDM is cheaper, and its melt processability is easier, but its elasticity and lower thermal resistance are its shortcomings in this field [16, 17].

The degradation of elastomeric materials in a simulated environment is commonly used to check their chemical stability in the fuel cell environment. Lin et al. [18] investigated the chemical degradation and dynamic-mechanical properties of five types of elastomers in the environment and functional conditions of fuel cells. They found fluorosilicone had the best chemical stability in the fuel cell environment, followed by EPDM, while liquid silicone and resin had cracks and the greatest color change. Liquid silicone and resin had a similar decrease in weight, while EPDM and FKM experienced only a slight increase in weight, and FSR did not experience any weight changes. In complementary research by the same researchers, experiments were conducted on these five types of elastomers, and the results indicated that resin, liquid silicone, and EPDM are better for sealing PEM fuel cells from a mechanical point of view, and in total, EPDM was introduced as the best rubber. Chao et al. [19] investigated the stress relaxation of EPDM rubber in a fuel cell and applied various environmental strains at different temperatures of deionized water. Their test results indicated that at up to a 25% strain rate, the stress-strain curve of this seals shows a linear behavior, and the yield modulus is only a function of time, regardless of the amount of strain. Shen et al. [20] tried to increase and improve the hardness and compression set of EPDM rubber using the multilayer method; they tested its soft and hard structure (based on the type of filler with which they vulcanized the rubber) in the form of alternating multilayers. Their results showed that the amount of the rubber's tensile strength and compression set in the multilayer structure was lower than the normal state; however, it adopted a higher hardness compared to the mixture of hard and soft rubber, and this hardness, unlike the compression set, increased with the increase of layers.

Wang et al. [21] investigated the chemical stability of silicone rubber under two types of pressure loading in a fuel cell environment. First, the surface morphology analysis showed that the initial smooth surface became uneven after being placed in the pile and this phenomenon resulted from the growth of removed cracks. Second, XPS and ATR-FTIR analyses showed that the surface's chemistry changed due to the loss of cross-links and some chemical bonds (because the fuel cell environment is an acidic environment at a temperature of 60 to 80 degrees). Finally, they came to the conclusion that the acid concentration inside the fuel cell environment as well as the amount of force loading (the force due to which the seal is under pressure) play an important role in the chemical and destructive stability of silicone rubber. Qiu et al. [22] selected some common rubbers, silicone (SR), fluoroelastomer (FR), nitrile (NBR), and EPDM, used for sealing fuel cells and applied chemical and mechanical analyzes. The results generally indicated that the pressure rate on the seals greatly impacts the seals' tensile performance. EPDM and SR rubber had the best and worst mechanical stability, respectively. From the temperature point of view, SR rubber had the most compatibility with temperature conditions up to 100 °C, and an increase in temperature caused the modulus of all seals to drop. Wu et al. [23] evaluated silicone rubber in the fuel cell environment and in cold temperature conditions (from -5, -10 and -20 degrees Celsius) and investigated its degradation and stabilization. They subjected the silicon to multiple temperature cycles and then assembled them into a fuel cell. With the help of various analyses, they showed that with the increase in the number of temperature cycles, the hardness of the rubber increased, while the sample experienced a greater weight loss. Chemical analyzes confirmed the previous research and showed that the surface chemistry changed and cracks are observed on the surface of the rubber sample.

In this paper, the chemical and mechanical properties of three different elastomers are investigated in the accelerated simulated environment of a PEMFC at room temperature and 80 °C. The results revealed that the molded EPDM has better compression test hardness, weight changes, tensile strengths, compression set, and spectrometry than the other analyzed elastomers.

2.Experiments

2.1. Materials and methodology

Some approaches to seal the PEMFC components include using flat gaskets, molded gaskets with desired profiles, integrated gaskets with MEA, and integrated gaskets with a bipolar plate. The flat gaskets are suitable for laboratory scale stacks, but this method needs more compression force than molded profiled gaskets for sealing. So, using the profiled gasket may help the montage procedure. In addition, the positioning of the profiled gaskets is much easier than the flat gaskets. Integrated sealing methods decrease the sealing surface number, causing better sealing than other methods. Integrating a gasket with MEA is difficult because of the sensitivity and thickness of the MEA. Choosing the material for the sealants is one of the most important parts of designing and manufacturing a fuel cell [24]. Durability is a vital parameter in commercial products. According to the US Department of Energy (DOE), PEMFC for mobile applications must operate reliably for at least 5,000 hours in a temperature range between -40 to 80 degrees Celsius [25]. To meet these needs, sealants chosen for PEMFC must also be sufficiently durable. Correctly designed sealants should ensure not only their efficiency in the initial stages but over the entire life of the fuel cells. Long-term laboratory tests often show that one of the limiting factors of the fuel cell life is leakage from the inside. This leakage or reduced performance of the sealants can be the result of long-term mechanical or chemical degradation.

The sealing force decreases due to the physical or chemical release of the material caused by material aging. Therefore, leakage can occur due to a reduction in the sealing force or the seal's failure [26-28]. During the fuel cell operation, the sealing material is exposed to mechanical stress in the presence of periodic temperature and chemical environments. The thermal cycle alone may increase the release of stress and pressure resistance, decreasing the clamping force of the fuel cell series and affecting the performance of the fuel cell [22, 29-31]. Degradation of the sealing material due to aging in a corrosive environment, temperatures up to 90 ℃, and mechanical stress may lead to loss of sealing power and thus cause external leaks, mixing of two gases, or short circuits of the plate. Since the long-term use of a fuel cell is highly dependent on the sealants' durability, increasing the resistance of the sealant material in the durability tests is a practical requirement to improve the life of the fuel cell. Therefore, it is very important to develop and manufacture durable seals that can last as long as the optimal life of a fuel cell.

2.2. Accelerated durability tests (ADTs)

In this set of tests, the changes in the properties of the selected materials are tested in an acidic environment and two temperature conditions [32-34]. The desired acid is a combination of sulfuric acid and hydrofluoric acid. The aging conditions of samples are listed in Table 1.

Table 1 . Aging Conditions

	Aging Solution	Aging Time duration	Aging Temperature
Sample	H_2SO_4 98%→1 M	0-120 day	Room temperature (23 °C)
	HF 48%→10 ppm	0-100 day	80 °C
	PH<1		

Since the pH value in a real fuel cell environment is 3-3.5, we tried to bring the test results closer to the real conditions by making a stronger acidic environment and reducing the test duration. The test was performed at room temperature (23 degrees) and 80 degrees in the oven. A temperature of 80 degrees was chosen to be closer to the working temperature of the fuel cell and to investigate the effect of temperature on the reaction rate between two materials (sealant and acid). The properties of the elastomers were measured before starting the test and recorded as the initial elastomer amounts. Then, at regular time intervals, the properties of elastomers were remeasured, and their changes compared to the initial value were calculated. The elastomer characteristics investigated in these tests were hardness, compression set, tensile strength, and weight change. The purpose of the tests was to investigate the durability of specimen materials in the desired fuel cell environment.

Three different sealants were chosen for the durability test. These sealants are EPDM, which is molded in a compression molding die; EPDM sheet, which is used for the sealant; and silicone, which is molded at room temperature.

A durability test was performed at room temperature for 100 to 120 days. During the test period, eight to ten samplings were done for each specimen on different days, and the amount of change in properties was recorded. Two types of samples were prepared for each selected material, a disk sample and a dumbbell sample. The disk sample was used for compression set tests, weight changes tests, and hardness changes tests and the dumbbell sample was used for tensile strength tests [34-36].

2.3. Standard tests

The following were performed to test the mechanical and chemical properties of the three selected sealants.

2.3.1. Hardness

Hardness indicates the elasticity of rubbers. The lower the hardness, the greater the elasticity of the rubber material. Hardness is defined as resistance to permanent indentation under certain conditions with a certain force. The hardness of elastomers can be classified in two ways: 1- inherent hardness and 2- hardness created during processes [37]. The Shore durometer standard comprises different classifications; the Shore A scale is used for rubbers, and the Shore D is used for plastics. Hardness is an essential parameter in determining the containment force and the amount of compression required for sealing. Excessive restraining force can cause plastic deformation of sensitive components such as the membrane and GDL, and low restraining force can increase the contact resistance between fuel cell components. Rubber hardness was measured using the AStM D 2240 standard test method [38-40] and the hardness test device shown in Fig. 1.

Fig. 1. Hardness test device.

2.3.2. Tensile strength

Sealants in fuel cells are exposed to compressive stresses; therefore, it is necessary to calculate the amount of stress applied to the sealants and compare it with the maximum allowed amount before compressing them. The tensile strength and hardness of an elastomer have a very close relationship with each other. The tensile strength of an elastomer largely depends on the ability of its crystals to strain during stretching. The AStM D 412 standard is usually used to determine the tensile strength of an elastomer [41]. The tensile strength test was performed by a two-ton universal tensile machine made by the Santam Company, as shown in Fig. 2.

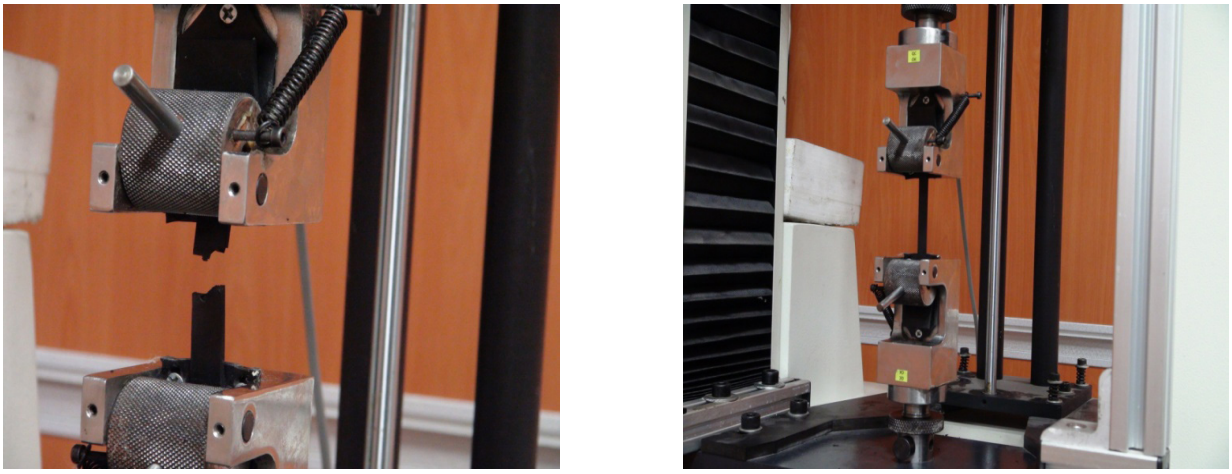

Fig. 2. Universal tensile machine made by Santam company.

2.3.3. Compression set

The compression set is actually the ratio of the elasticity to the viscosity of an elastomer against compressive force. The compressive modulus indicates the ability of an elastic material to return to its original dimensions or maintain elastic properties after compression for a long time. This test uses the standard AStM D 395 (ISO 815) and constant strain method. The mechanism required for the compression set test was designed and built according to the mentioned standard [42]. This mechanism, shown in Fig. 3, includes two steel plates with dimensions of 10×10×2 cm and two 9.5 mm thick bars [43-45].

Fig. 3. Mechanism designed for compression set.

Three samples were evaluated in each compressive modulus test, and their average was considered the elastomer compressive modulus value. The compression set for each sample is calculated as follows.

$$C_B = \frac{t_0 - t_i}{t_0 - t_n} \times 100 \quad (1)$$

where, t_o is the initial thickness, t_i is the final thickness, t_n is the thickness of the spacer bars, and C_B is the compressive modulus of the elastomer.

2.3.4. Chemical resistance

Chemical resistance indicates the degree of stability of a material in a certain environment, and it is manifested in the form of changes in the physical and chemical properties of the material, such as weight, hardness, tensile strength, etc. The test is conducted by measuring the material properties before the test, then the material is exposed to the simulated environment at a certain temperature and for a specific period of time, and then the properties are measured again. The amount of changes indicates the level of stability. Small changes indicate the compatibility of elastomer, and large changes indicate a lack of compatibility [46]. The elastomer is placed in a simulated fluid for a specific time, and then changes in dimensions, weight, and other physical properties are measured and recorded as a percentage of the initial value per the AStM D 471 standard. After removing the samples from the simulated environment, first, they should be washed with water to clean their surface from any acidic pollution; then placed at room temperature for 2 hours, and finally, the necessary properties measured. One of the important properties examined in this test is the change in sample weight [47-49]. Before placing the disks in the simulated acidic medium, they are weighed, then after a certain period of time, they are weighed again. The amount of change is expressed as a percentage of the initial weight. Eq. (2) shows the weight change percentage calculation.

$$C = \frac{w_2 - w_1}{w_1} \times 100 \quad (2)$$

In this relationship, w_2 is the final weight of the sample, w_1 is the initial weight, and C is the weight change percentage. Fig. 4 shows the digital scale used to test weight changes.

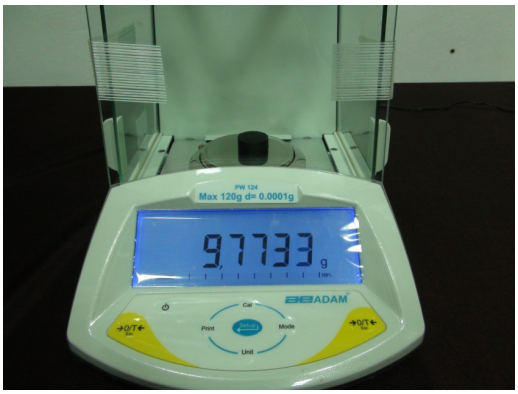

Fig. 4. The device used in the weight change test.

2.3.5. Spectroscopy

An important sealant feature is the stability of its chemical compounds in the fuel cell environment. Since the seal in the fuel cell is placed in an acidic, oxidizing, and humid environment, the stability of the sealant's chemical composition in this environment can indicate its suitability for use in fuel cells [50-52]. Changes in the chemical composition of the sealant and other components can be detected with the help of spectroscopy. Spectroscopy is the study of matter and its properties by examining light, sound, and particles emitted, absorbed, or scattered from the matter. There are two types of spectroscopy: atomic absorption spectrometer and Fourier transform spectrometer. An atomic absorption spectrometer is usually used to check the leach of new elements from the sealant in

the acidic solution. However, the Fourier transform spectrometer is used to investigate any changes in the chemical composition of the sealant. Atomic absorption and Fourier transform spectroscopy was carried out at a standard research center.

3. Results and discussion

Two types of disk and dumbbell samples were prepared for each selected specimen. The disk sample was used for compression set tests, weight changes, hardness, and spectroscopy, and the dumbbell sample was used for the tensile strength test. EPDM disk and dumbbell samples were cut from the sheet according to AStM D 395 standard [53]. In addition, two other specimens were molded. According to Fig. 5, the silicone rubber sealant specimen needed to be molded. Silicone was poured into prepared Teflon molds to make the disk and dumbbell-shaped samples because it has no adhesion with silicone adhesive. For proper processing, the samples should be left in the open air for 4-5 days. Then they can be separated from the mold. During the molding, the silicone should be poured in such a way that no air bubbles are created between the samples. The basic specifications of this item are listed in Table 2.

Table 2. Basic Specifications of the Silicon Seals

Material	Molded EPDM	EPDM sheet	Silicon rubber
Density (g/cm^3)	1950	1850	1700
Hardness (Shore A)	59	64	42
Tensile strength (MPa)	9.5	6.2	2
Compression set (%)	3	16	28

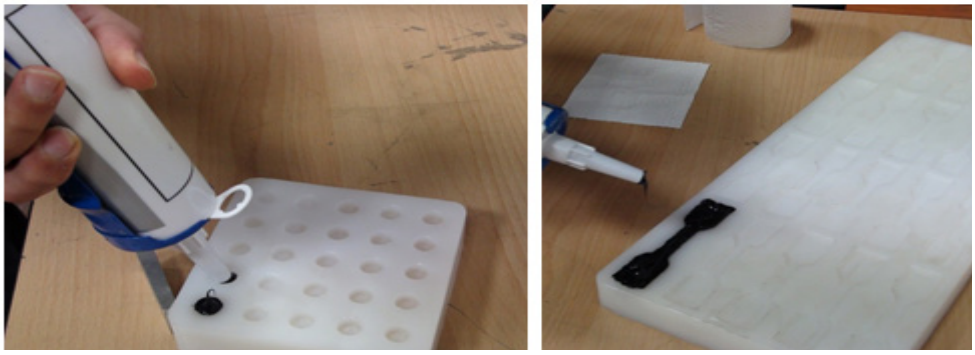

Fig. 5. Silicone rubber molding of the disk and dumbbell-shaped samples.

After preparing the samples and measuring their basic properties, they are placed in special autoclave containers that can withstand temperatures up to 150 degrees. As shown in Fig. 6, some of these samples are placed in the oven, and other are left at room temperature.

Fig. 6. Samples placed in the oven and at room temperature.

Sampling was done for each specimen at specific time intervals, and the amount of change in the properties of each sample was measured. The results of the hardness test at room temperature (23 °C) and in the oven (80 °C) are shown in Fig. 7a and Fig. 7b, respectively. As shown in these figures, molded EPDM and silicone changed very little, but the EPDM sheet had a continuous decreasing trend throughout the test period, decreasing from 63 to 45 Shore A. The molded EPDM hardness did not change much at room temperature, but its hardness increased slightly, increasing from 59 to 65 at 80 °C. The EPDM sheet had the most harness changes in these tests at both room temperature and 80 °C.

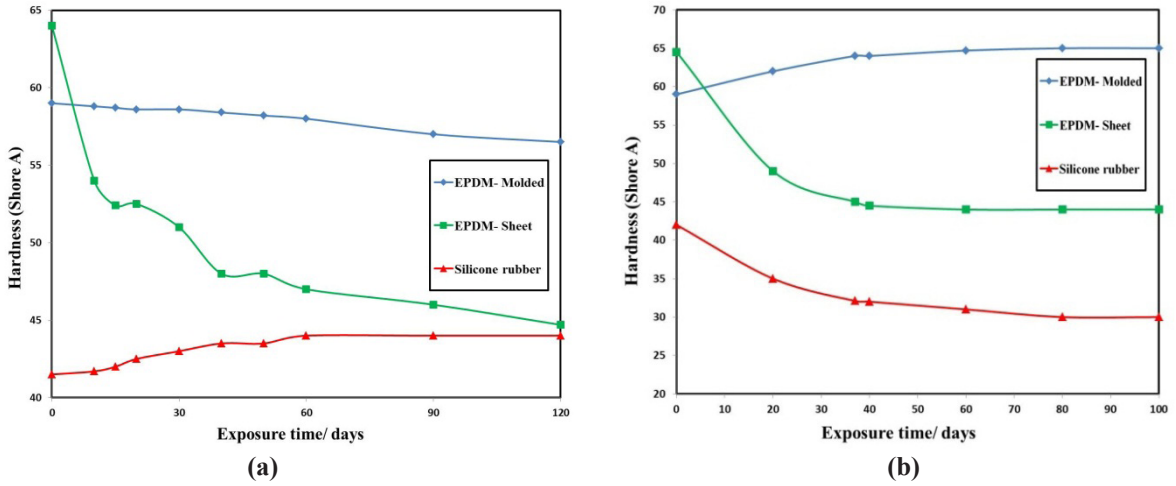

Fig.7. Changes in the hardness of sealants (a) at room temperature and (b) at 80 °C.

Figs. 8(a) and 8(b) show the tensile strength changes at room temperature and in the oven, respectively. Silicon had the highest percentage of changes in the experiment at room temperature. The tensile strength of the silicone changed from 2.3 Mpa to 1.4 Mpa after 120 days. EPDM molding showed an insignificant change in tensile strength tests. The EPDM sheet also had a smooth continuous decreasing trend.

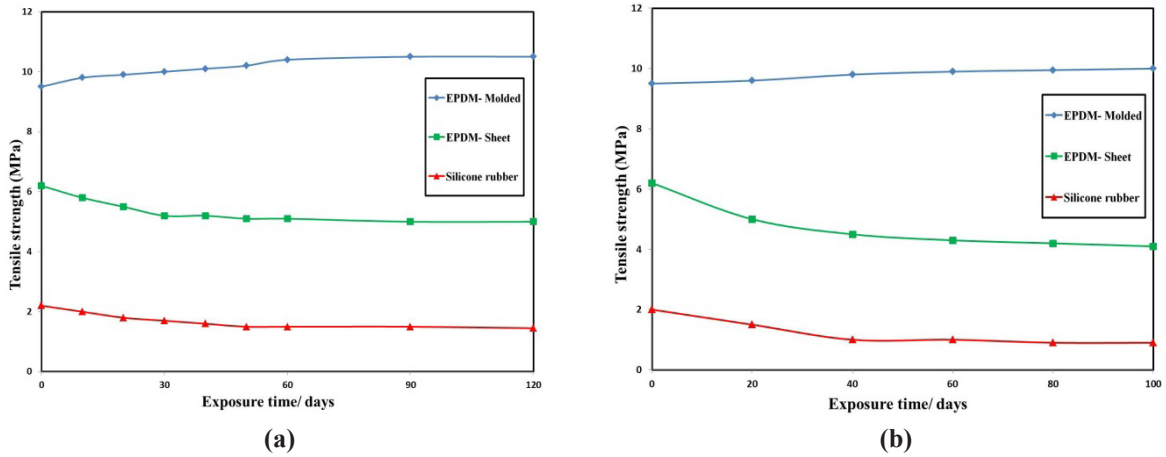

Fig. 8. Changes in tensile strength for sealants (a) at room temperature and (b) at 80 °C.

Figs. 9(a) and 9(b) show the results of compression set tests at room temperature and in the oven, respectively. Molded EPDM had a big initial change, increasing from 3% to 7%. However, after this, the amount remained almost constant. In this case, the silicon showed the most amount of change.

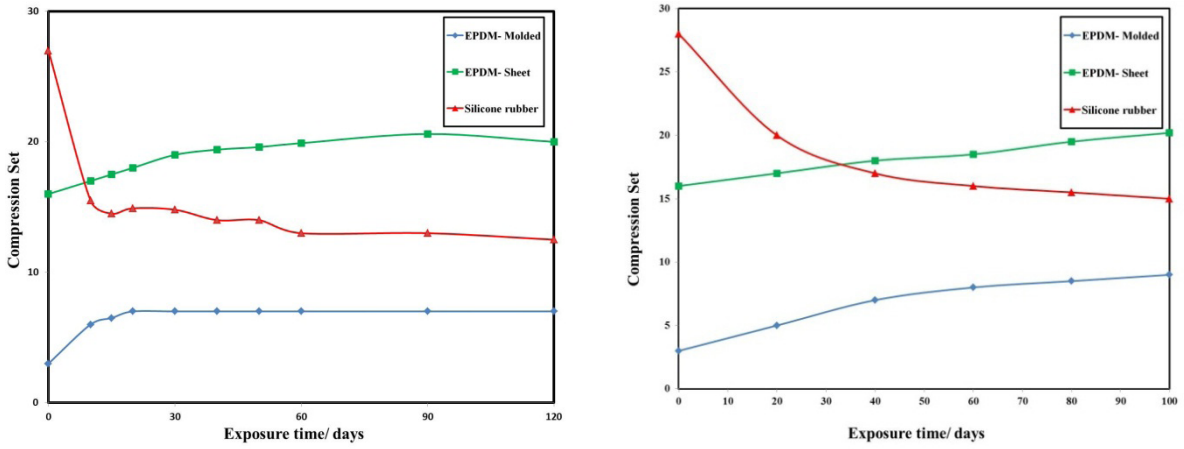

Fig. 9. Changes in the compression set for sealants (a) at room temperature and (b) at 80 °C.

The samples' water absorption results are shown in Figs. 10(a) and 10(b) at room temperature and in the oven, respectively.

As shown in these figures, the silicone specimen's weight increased, and the EPDM molding weight decreased in both tests. The percentage of silicone changes is more than that of molded EPDM changes, and the amount of changes in both materials in the oven is more than at room temperature. In other words, in both tests, elements are separated from EPDM and enter the solution, while silicone absorbs elements from the solution, which increases its weight. The weight changes created in EPDM molding in both tests were very small.

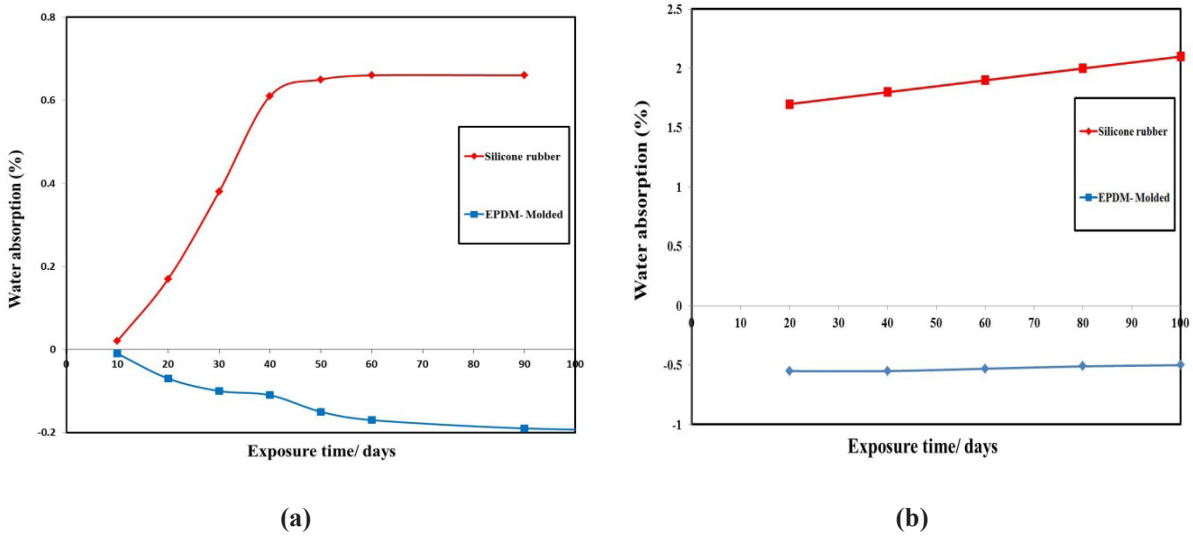

Fig.10. Weight changes in the sealants (a) at room temperature and (b) at 80 °C.

Because the obtained results from the EPDM sheet and silicone rubber in previous tests showed that they were not stable in the fuel cell accelerated simulated environment, Fourier transform spectroscopy was performed only on molded EPDM, which had better durability in previous tests. The results of the spectroscopic experiment at room temperature and in the oven are shown in Figs. 11 and 12, respectively. The results obtained from this test show insignificant changes in the bonds in the molded EPDM after the test. Almost all bonds remain in the chemical composition, and only the intensity of some was reduced. These results confirm the previous tests and show the stability of this material in the fuel cell environment.

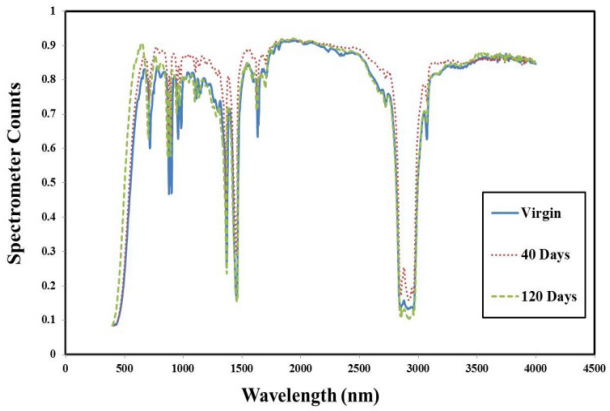

Fig. 11. Spectroscopy plot of molded EPDM sealant at room temperature.

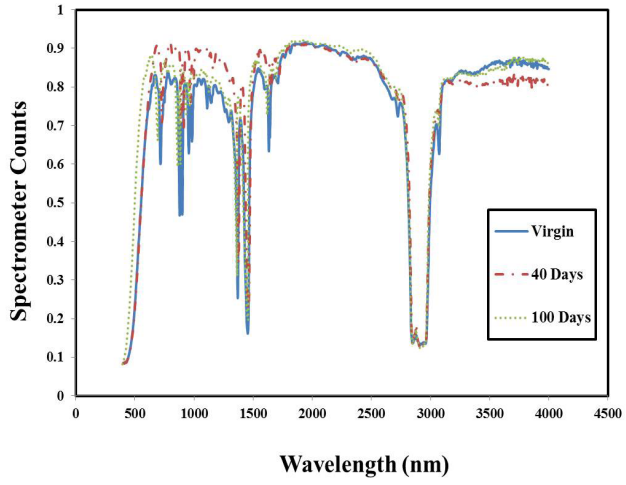

Fig. 12.Spectroscopy diagram of molded EPDM sealant at 80 degrees

Atomic absorption spectroscopy was performed only for the acid samples in which the molded EPDM gasket was placed. The results obtained from this test confirmed the correctness of the weight changes test. Calcium and magnesium are the elements whose concentration was studied in the acidic sample. The results obtained from this test which is shown in Table 4 indicate that magnesium does not separate from the surface of the sealant at room temperature, and increasing the temperature causes some of it to separate and dissolve in acid. Also, calcium is separated from the sealant in a small amount at room temperature and dissolves in acid. But increasing the temperature increases the dissolution rate of calcium in acid. Despite all the mentioned contents, the amount of elements added to the acid is not so high that it shows a large change in weight, and the weight loss of molded EPDM has been very small.

Table 4. Atomic absorption test results.

Samples	Mg (PPM)	Ca (PPM)
prototype	1.2	2.3
After 1000 hours at 23 °C	6.1	1.2
After 3000 hours at 23 °C	15	2.5
After 1000 hours at 80 °C	14	2.4
After 2500 hours at 80 °C	47	6.6

Fig. 13 shows the silicone sealant before and after the test. As it is clear in the figure, holes and cracks have been created on the surface of the silicon after 100 days in the acidic environment of the fuel cell at an oven temperature, indicating the silicon's degradation. However, the changes in the appearance of the EPDM sheet were much less compared to silicone; the molded EPDM also had very few changes compared to the previous two materials. However, the large changes made in the silicon surface should be noted in this experiment, which confirmed the results obtained from the previous experiments.

Fig.13. Changes in the appearance of the silicone sealant.

4. Conclusion

In this research, the mechanical behavior of the changes in the properties of three selected fuel cell sealant materials, EPDM sheet, EPDM molded, and silicone rubber, has been investigated. Mechanical properties such as weight change percentage, tensile strength, compression set, spectroscopy, and hardness have been evaluated for these three materials. An accelerated process that simulated the harsh environment of the fuel cell was used to investigate the mechanical and chemical properties of the sealant at the ambient temperature and working temperature of the fuel cell during specific and predetermined times. Results revealed that the working temperature of the fuel cell causes more changes in mechanical properties compared with the ambient temperature. The comparison of the results shows that silicone has the most properties changes of the three types of sealant, and molded EPDM was the least influenced when exposed to the two temperatures. Also, the results obtained in all tests show the durability and stability of molded EPDM in the acidic environment of fuel cells. Considering that component durability is one of the requirements of fuel cell industrialization, using this material in these projects seems sensible.

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