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Effects of curing systems on the mechanical and chemical ageing resistance properties of gasket compounds based on ethylene-propylene-diene-termonomer rubber in a simulated fuel cell environment

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ABSTRACT

Ethylene-propylene-diene-termonomer (EPDM) rubber based fuel cell gasket compounds have been designed and explored the effects of various vulcanization systems on different properties. Three types of sulphur-accelerated vulcanization systems such as conventional vulcanization (con), semi-efficient vulcanization (sev) and efficient vulcanization (ev) and also a peroxide vulcanization system were employed in this study. The curing characteristics, tensile, hardness and compression set properties of the cured compounds were evaluated. The crosslink density was assessed by equilibrium swelling method in dodecane. The chemical stability of the cured EPDM compounds was also evaluated through an accelerated durability test (ADT) using a solution (1 M H_2SO_4 + 10 ppm HF) very close to the fuel cell atmosphere. Attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR), energy dispersive X-ray spectroscopy (EDX) and scanning electron microscopy (SEM) were employed to investigate the chemical and physical changes of the cured EPDM compounds before and after exposure to the ADT solution over time. The results indicate that the EPDM compounds cured with peroxide exhibit the highest crosslink density with lowest compression set value at both room temperature and at elevated temperature. The FTIR and the corresponding SEM results show no significant chemical degradation of the peroxide cured EPDM compounds due to ADT ageing.

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Introduction

A fuel cell is an electrochemical device which converts the chemical energy of a fuel directly into electrical energy. The proton exchange membrane (PEM) fuel cell is one of the most common types of the fuel cells. Recently, it has gained special attention for various applications since it can yield high power density without any environmental pollution [1–5] A PEM fuel cell consists of membrane electrode assemblies stacks (MEA) including the gaskets sandwiched between bipolar plates as illustrated in Fig. 1.

Elastomeric materials are often used as gaskets and seals in PEM fuel cells to prevent the reactant gases from leaking or mixing each other directly during operation or repose. The gaskets in PEM fuel cells are always being exposed to an acidic liquid solution, humid air as well as being subjected to compressive stress between the bipolar plates. It has to withstand all of these extreme conditions at the operating temperature (60–120 °C) for several thousand hours. The failure of any single gasket in the PEM assembly during either operation or rest may lead to mixing up the reactants which will adversely affect the overall operation and performance of the fuel cell. Therefore, it is very essential to understand the failure behaviour of the fuel cell sealing gasket in a long run which is generally evaluating by conducting accelerated ageing studies in a simulated condition which is very close to the actual fuel cell working atmosphere [6–10].

The major requirements for an elastomeric fuel cell gasket compounds are long term stability in acidic atmosphere, good stability in humid air and hydrogen, superior thermal stability, high mechanical strength with low compression set at low to high temperature, high electrical insulation property and adequate hardness with good processability. Moreover, the gasket materials must be free from ingredients that might be eluted to block the pores of the gas-diffusion layer or to coat other active surfaces of the fuel cell components. Many elastomeric materials have been explored as candidate materials for PEM fuel cell gaskets [11–13]. Fluroelastomers, silicone rubbers and hydrocarbon based elastomers such as EPDM are common choices for the developments of fuel cell gaskets compounds because of their superior chemical and thermal resistance properties. However, there are problems associated with each system. For instance, fluoroelastomers offer excellent chemical resistant against acids and water, and sustained hardness with very low compression set and hence expected to do work as an excellent sealing material with good durability for PEM fuel cell applications. As yet, fluoroelastomers are not a primary choice by the manufacturers because of its poor melt processability for injection moulding and it also has got poor low-temperature flexibility (Tg of FKM is around −20 °C). Moreover, they are much more expensive than the other general purpose rubbers [14–16]. Silicone rubber offers good thermal resistance, but it has a high compression set value, poor chemical resistance under acidic environment and also more expensive than other commodity rubbers [17–19]. EPDM is an outstanding polymer with good resistance to heat, light, ozone, and UV exposure and is widely used for outdoor applications. Even though EPDM is a very cost effective rubber with good melt processability, it has the disadvantages of lower elasticity and thermal resistance compared with fluoroelastomers [20,21].

Many research reports are available in the literature with a special attention to the chemical degradation behaviour of the above mentioned elastomeric materials which are being considered as potential candidates for the fuel cell gaskets. For example, Tan et al. have investigated the mechanical and chemical degradation characteristics of a silicone rubber based gasket compound in a fuel cell environment. Their investigation revealed that silicone rubber based gasket compound undergo severe degradation in the simulated fuel cell environment as a result of which elements such as silicon, calcium and magnesium are leached out from the gasket which adversely affect the electrochemical process in a fuel cell system [22–24]. They have also reported the mechanical property degradation of EPDM and FKM in a simulated acid solution which is very close to the actual PEM fuel cell environment using micro-indentation and dynamic mechanical analysis [9,10,25]. Mitra et al. have investigated the chemical degradation of both the sulphur/accelerator crosslinked and peroxide/coagent cured EPDM rubber in a 20% Cr/H_2SO_4 acidic environment. From their investigation, they have concluded that both the sulphur/accelerator cured EPDM as well as the peroxide/coagent cured EPDM undergoes chemical

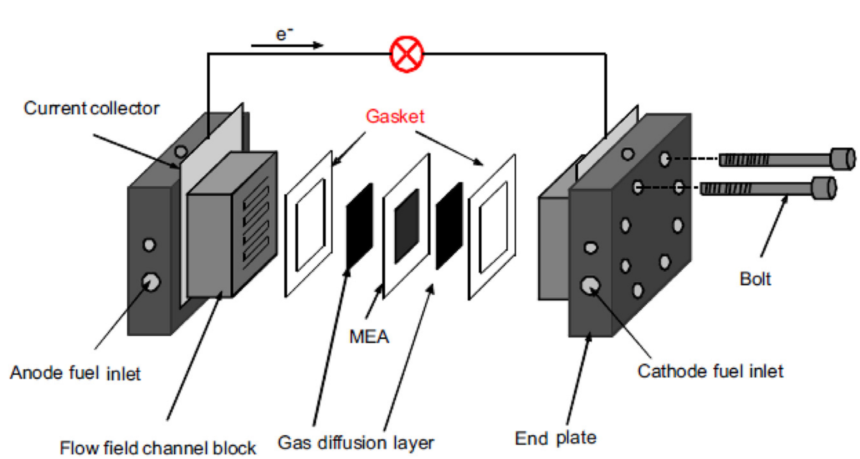

Fig. 1 – A typical schematic diagram of PEM fuel cell with its various components.