

heteropolyacid has been used successfully as promoter in the catalyst surface to increase stability to CO poisoning, and has the necessary characteristics to serve as a mediator for reductions<sup>(7-13)</sup>. We observed that Silicate and PWA can increase chemical and temperature stability, more retain water molecule and increase flexibility of the membrane so that the composite membrane could be operated at temperature above 80°C in low humidity without shrinkage<sup>(6)</sup>.

#### Material and Method

Composite membrane was prepared by<sup>(12)</sup> by hot pressing the mixture of melt fabricable perfluorosulfonylfluoride copolymer resin (Nafion) with fine zeolite powder at 230°C

In these investigations an embedded Si/PWA in the polymer matrix using sol-gel method to produce hybrid membrane was performed. The membrane was embedded through two steps, first heteropolyacid (PWA) was mixed with Tetra Ethoxy Ortho Silicate (TEOS) to form solution at room temperature. Ratio of SiO<sub>2</sub>/PWA used in this experiment is 10:3 (w/w)<sup>(13)</sup>. TEOS in acid condition will be hydrolyzed produce silanol (Si(OH)<sub>4</sub>). Second step was entrapped both component in the polymer matrix during gelling reaction of the silicate that go on in situ during drying in vacuum oven at temperature 80°C 25 mmHg and during hot pressing at 30 atm and 120°C and PWA is entrapped in the silica matrix polymer.

Immobilizes PWA in a matrix porous have been conducted by<sup>(9)</sup>. Zeolite is used as matrix (super cage) to immobilize Phosphotungsticacid (PWA) by arranging ratio aluminum/silicate molecule of zeolite. In this process required cation for ion exchange and solvent to zeolite dealumination hence the process is expensive and is not simple.

To overcome this problem we choose the process to immobilized PWA in the silica matrix using sol-gel method. After PWA has been immobilized, than entrapped both material in the membrane matrix polymer using impregnation method to produce hybrid membrane (Nafion/Si/PWA). Membrane will use as solid electrolyte for the Proton Electrolyte Membrane Fuel Cell (PEMFC) at high temperature (above 80°C).

The whole procedure to modifies membrane electrode assemblies (MEA) using sol-gel method conducted by few step that are: a). Impregnation Nafion membrane using solution contain (TEOS, PWA, IPA, water). b). Drying impregnated membrane at room temperature. c). Casting the catalyst ink solution on both surface of the membrane to produce membrane-electrode. d). Drying membrane-electrode at room temperature. e). Drying membrane-electrode in vacuum oven at 25 mm Hg and 80°C during 30 minute produce dry membrane-electrode. f). Superimposed GDL on both side of the membrane-electrode and than hot press at 120°C and 30 atm during 4 minute. g). Test of the MEA using Arbin fuel test system to know activity of the catalyst. Scanning electron microscopy (SEM) to analyze the microstructure of the catalyst layer.

#### Gas diffusion layer (GDL) preparation

Diffusion layer forming composition was prepared as follows.

The first Vulcan (carbon black) a mount of 1850 mg with 56.37 g Iso Propyl Alcohol mixed using magnetic stirrer during 30 minute. The resultant mixture was added Polytetrafluoroethylene 666 mg (PTFE wt 60%) mixed using homogenizer for 30 minute. The diffusion layer forming composition was applied onto the carbon cloth and dried at vacuum oven at temperature of 120°C for 2 hour. The finally obtained diffusion layer exhibited carbon loading 4,17 mgcm<sup>-2</sup>.

#### Catalyst layer preparation.

Catalyst layer forming composition was prepared as follow.

1826 mg 20 wt% Pt/C catalyst and 5.699 g deionized water (DI) were mixed and stir using mechanical stirrer for 3 minute. To mixture of the catalyst and DI water was added 2113 mg Nafion ionomer produced by (Du Pont) solution 10 wt%. Than mixed using magnetic stirrers for 5 minute. To the mixture added 3 gram of Iso Propyl Alcohol (IPA), mixed using magnetic stirrer for 3 minute. The finally obtained Pt loading on the surface is 0.38 mgcm<sup>-2</sup>.

#### Membrane Electrode (ME) and preparation Membrane Electrode Assemblies (MEA).

##### Treatment of Nafion membrane.

Nafion 112 were treated to remove organic and metal impurities by refluxing in the 3% wt (H<sub>2</sub>O<sub>2</sub>) peroxide solution for 1 hour. Followed by refluxing in the boiling water for 1 hour. To remove trace organic and metal impurities membrane treated again in the 1 N sulfuric acid solution (H<sub>2</sub>SO<sub>4</sub>) for 1 hour. The membrane was then refluxed in the boiling water 3 times until pH of water was neutral.