Graphene has been considered as an ideal material for flexible gas barrier layer since it is impermeable to most of the gaseous species as well as is flexible [1]. Graphene oxide, which is readily soluble in various solvents, has been layered with polyelectrolyte film using layer-by-layer (LBL) process and studied as a possibilities of the gas barrier layer [2]. The graphene oxide/polyelectrolyte multilayer thin film reduced the permeability of H₂O by three orders of magnitude compared with bare PET film, but was not sufficient for the application as the barrier layer for OLED applications.

There are several parameters that may have contributed to the higher permeability of moisture than expected. Firstly, the graphene oxide/polyelectrolyte layer are processed in aqueous solution and the barrier layer is, therefore, hydrophilic in nature. With the hydrophilic nature, the solubility of H₂O in the barrier layer would be high, increasing the permeability of the moisture. Geim et al [3] demonstrated that the permeability of graphene oxide layer can be reduced dramatically, almost by seven orders of magnitude, by reducing the graphene oxide, making hydrophobic. Secondly, the thickness of polyelectrolyte is ~3nm thick and that may have reduced the wetting angle of H₂O and also increased the permeability of the moisture.

Based on above premises, two different approaches were adopted in this study. Firstly, the polyelectrolyte and graphene oxide layers were converted to hydrophobic layer by heat treatment. The polyelectrolyte materials were selected such that they became converted to hydrophobic in nature upon heat treatment. Secondly, graphene oxide/polyelectrolyte multilayer was prepared, in which the polymer layer is ~10~100 nm thick. Fig. 1 shows the SEM micrographs of graphene oxide coated on polystyrene thin film. Graphene oxide was coated the film uniformly and its area fraction was better than 99%. Moisture permeability of the reduced graphene oxide/polymer multilayer coating are being characterized currently and will be presented at the meeting.

In a different approach, the polyelectrolyte layer used in the LBL processing was converted to hydrophobic layer via heat treatment. Two different cationic and anionic polyelectrolyte were used in this study. For the cationic polyelectrolyte, PAH was used and for the anionic polyelectrolyte was used. After the LBL coating of multiple dyads consist of graphene oxide/PAH, the coating was heat treated at 180°C for 2 hour. to convert the carboxyl ligands to hydrophobic ligands. Using the multilayer barrier coating, moisture permeability was measured using Ca-test method as shown in Fig. 2. As shown in Fig. 2, the reduction of graphene oxide and conversion of polyelectrolytes have enhanced the barrier characteristics up to \(10^4\) g/m²·day, but the lag time is rather short. As the lag time is mainly determined by the thickness of the barrier layer, it is expected to have longer lag time with the graphene oxide/polymer multilayer barrier film prepared in the proceeding section.

Fig. 1. Polymer/Graphene oxide SEM image

Fig. 2. Electrical conductivity of Ca at 40°C 85% RH

References