Opinion

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Decarboxylation Induced Crosslinking: a Promising Method to Improve the Anti CO_2 Plasticization of Gas Separation Membranes

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Natural gas containing CO_2 will cause pipeline corrosion and decrease calorific capacity value. In the past, traditional methods, such as adsorption, cryogenic distillation, pressure swing adsorption in some applications were used for CO_2 removal [1]. Concerning cost, requirements for energy and effectiveness, membrane separation is a better choice for natural gas sweetening. Membrane materials are noticeably important for achieving high gas permeability and selectivity. In the case of CO_2/CH_4 separation, the ability of anti CO_2 induced plasticization is especially important. Until now, a number of polymers are available to be used as membrane material such as: cellulose acetate, polysulfone, polydimethylsiloxane, polycarbonate, polyimide etc. [2]. Among them, aromatic polyimides are very prospective due to their characteristic of high thermal stability, good mechanical strength, and chemical stability. Polyimide membranes have been used in applications such as the separation of O_2/N_2 and CO_2/CH_4 [1]. However, due to the limited resistance to CO_2 plasticization, polyimide membranes need to be further modified.

In order to solve the problem, most of researches focus on the crosslinking of polyimide using either chemical crosslinking or thermal crosslinking protocol. Chemical crosslinking uses cross-linking agent to link polymer main chains. However, the crosslinking bonds such as ester or imide groups may suffer from hydrolysis when the feed streams consist of acid gas or high temperature operation which will result in degradation of cross-linking bonds [3,4].

To improve the chemical stability of the crosslinking group, Koros' group prepared the polyimides containing carboxylic acid by thermally induced decarboxylation reaction to crosslink. The polyimides were crosslinked by more stable C-C bonds and had stable CO_2/CH_4 selectivity at CO_2 pressure of 30 atm [5]. Based on their research, our group synthesized two carboxylic acid-containing diamines CADA1 and CADA2 as well as prepared 6FDA-CADA1 and 6FDA-CADA2, respectively. The decarboxylation crosslinked polymers also showed the improved CO_2/CH_4 separation properties which were stable at CO_2 pressure up to 30 atm. However, the thermal decarboxylation crosslinking reactions were completed at the 425°C which was above the Tg of polymers. This would lead to pore collapse of the polymer if it was fabricated into an asymmetric membrane. To address this issue, we synthesize two phenolphthalein-based polyimides, 6FDA-MPP and 6FDA-PP using an alternative method of thermal oxidative crosslinking at 375°C. The results indicate that the temperature of thermal oxidative crosslinking process is lower than thermal induced decarboxylation crosslinking (this work has been submitted).

Recently, inspired by the structure of 6FDA-Durene: DABA (3:2), we replaced DABA monomer with CADA1 and synthesized the 6FDA-Durene-CADA1 copolyimides. Previous research in our group indicated that CADA1-based crosslinking polyimide had higher inter-chain distance due to the presence of a biphenyl group between the carboxylic acid group and the main polymer chain [3]. So we expected that the 6FDA-Durene-CADA1 polyimides should have improved gas permeability and selectivity than 6FDA-Durene-DABA.

Reference

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