

## CHAPTER I

### INTRODUCTION

In natural gas, methane is the major component and also contains a certain amount of carbon dioxide. The existence of carbon dioxide decreases the heat value of natural gas remarkably. Furthermore, in a moist environment, i.e. carbonic acid can be produced by water and carbon dioxide, which corrodes not only the equipment, but also the pipes in the transportation. Recently, some researchers think that if carbon dioxide is collected and injected into the oil field, it will promote the production of the oil (Zhang *et al.*, 2010). Therefore, removing of carbon dioxide from natural gas is necessary.

Polymeric membrane, one of attractive techniques for gas separation, can provide advantages in terms of low capital investment, low energy consumption, high process flexibility, energy efficiency, and ease of operation. However, the performance of pure polymers is limited by an upper-bound trade-off line, which demonstrates a strong inverse relationship between permeability and selectivity (polymers that are more permeable are commonly less selective and vice versa) (Zornoza *et al.*, 2011). To improve performance, new materials and procedures for membrane fabrication are being investigated. One potential strategy for fabricating high performance polymeric membranes involves in the introduction of inorganic fillers into polymers, forming so called mixed matrix membranes (MMMs) (Kim and Marand, 2008; Xing and Ho, 2009). The formation of an inorganic–organic material with excellent interfacial compatibility between phases is the most prominent challenge in hybrid membrane performance since the advantages of both phases are complemented each other: high selectivity of dispersed fillers and desirable mechanical properties of polymers.

In this study, polybenzoxazine is selected as the continuous phase because of its high performance thermosetting resin, which exhibits excellent properties, such as low shrinkage after curing, low water absorption, good thermal stability and high glass transition temperature.

From the previous work (Ployangoonsri, 2010), mixed matrix membranes, comprising of polybenzoxazine and ZSM-5. were prepared. The MMMs resulted in

the drop-off CO<sub>2</sub> and CH<sub>4</sub> permeability, as compared to the pure polybenzoxazine membrane. Moreover, increasing the zeolite loading did not significantly improve either gas permeability or CO<sub>2</sub>/CH<sub>4</sub> selectivity since molecular sieving mechanism of ZSM-5 plays a minor role in CO<sub>2</sub>/CH<sub>4</sub> separation. Recently, mesoporous molecular sieves have been used in MMMs to enhance permeability or selectivity, for instances, Kim *et al.* (2005) enhanced gas permeability of polysulfone (PSF) by incorporating mesoporous MCM-48, Zhang *et al.* (2008) reported Matrimid<sup>®</sup> MMMs with calcined MCM-48 for gas separation. They showed that mesoporous materials increased the permeability of the MMMs without decreasing selectivity due to good compatibility with the polymer matrix.

The mesoporous MCM-48, to be used as filler in polybenzoxazine, has attracted considerable attention because it has a three-dimensional interconnected cubic pore structure, high surface area, ordered pore structure array, and narrow pore size distribution. This research focuses on the preparation and characterization of mixed matrix membranes, composing of polybenzoxazine as the matrix and mesoporous MCM-48 as the molecular sieve. The permeability/selectivity properties are tested for the gas separation of CH<sub>4</sub>/CO<sub>2</sub>.