

Application of carbon molecular sieve membranes in a mixed hydrogen-natural gas distribution network

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1. Summary

Carbon membranes have been identified as a promising technology for separation of hydrogen from natural gas. These are produced by carbonization of cellulose in vacuum or in an inert gas atmosphere. Mixed gas tests show a hydrogen selectivity over methane greater than 1000 and a pressure-normalised flux greater than $0.2 \text{ m}^3(\text{STP})/\text{m}^2\cdot\text{bar}\cdot\text{h}$. Carbon membranes may experience a loss of permeability when exposed to strongly adsorbing gases at ambient to moderate temperatures. Electrothermal treatment is presented as a possible solution to this aging issue. A procedure for making hollow carbon fibres is also presented.

Keywords: H₂, membranes, carbon, natural, gas

2. Extended Abstract

The transition to a full hydrogen distribution system may be a lengthy and costly exercise; hence a transitional approach using existing natural gas (NG) networks to transmit mixtures of hydrogen and NG is being investigated. NaturalHy, a 6th Framework Programme EU project, in which the NTNU is a participant, aims to test all critical components in a mixed network by adding hydrogen to existing natural gas networks.

Membranes have been identified as a promising technology for recovering hydrogen for end-use. One promising membrane material, under development at the Membrane Research Group (MEMFO) at NTNU, is the carbon molecular sieve. This is produced by the carbonisation of cellulose at temperatures of 550-750°C to produce a nanoporous carbon film, capable of discriminating between the smaller hydrogen molecules and the remaining molecules in a NG stream. The starting material, cellulose, is also cheap and abundant.

Carbon membranes at MEMFO are at the bench scale testing stage. Performance has been shown to depend on the temperature, pressure and composition of the feed. Mixed gas feed tests show that carbon membranes can achieve a hydrogen selectivity over methane greater than 1000 and a pressure-normalised flux greater than 0.2 m³(STP)/m².bar.h.

An obstacle for a breakthrough of carbon membranes has been the loss of permeability when exposed to strongly adsorbing gases at ambient to moderate temperatures. This paper addresses a possible solution to this aging issue. When a low voltage direct current is applied to the metal doped carbon during a permeation test, the permeability increases instantaneously and remains stable. The relative permeability increase depends among other factors on the gas type and on the amount of current applied. Carbon membranes containing different metals will be compared with respect to the effect of electrothermal treatment. Possible explanations of the phenomenon will also be given. A great advantage of this regeneration technique is the on-stream operation, avoiding shutdown or switching to an extra set of membranes.

To be commercially attractive, the packing of the membrane inside a module has to be efficient. Hollow fibres provide a configuration with a packing density up to 30 000 m² membrane per m³ module. A procedure for making such fibres from cellulosic materials will be presented.

Membranes placed at various end-user sites on the distribution network must cope with varying hydrogen concentrations in, and pressures of, the feed. However, process simulations show that 80% of the hydrogen can be recovered from the pipeline or a slipstream thereof at a purity greater than 90 vol%, in a single separation stage. The volume and footprint of these membranes would be small and they could conveniently be placed at commercial and residential sites.