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Crosslinking polyimides for membrane applications: A review

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ABSTRACT

This review discusses many crosslinking methods for polyimide membranes that have been described in literature. Some important properties of polyimides and their synthesis reactions are first summarized. The important (commercialized) polyimide types that are now used in membrane technology, the preparation methods available for polyimide membranes and their main applications are listed. The effects of thermal annealing of polyimide membranes are briefly discussed, before giving an extensive review of the many crosslinking methods that have been described in literature. Thermal crosslinking, UV crosslinking and a range of chemical crosslinking methods, including diol and diamine crosslinking, are discussed in detail, focusing on the actual chemistry behind the crosslinking. Also, some new, not yet fully studied, crosslinking methods are listed.

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Abbreviations: [BMPyr][Tf2N], 1-butyl-1-pyrrolidinium bis(trifluoromethylsulfonyl)imide; [MOct3N][Tf2N], methyltrioctylammonium bistriflimide; 4-MPDA, 3,5,6-tetramethyl-1,4-diphenylenediamine; 6FDA, hexafluoroisopropylidene-diphthalic anhydride; BAPP, 2,2-bis[4-(4-aminophenoxy)phenyl]propane; BTDA, 3,3',4,4'-benzophenonetetracarboxylic dianhydride; CL, crosslinking, crosslinked; CTC, charge transfer complex; DABA, 3,5-diaminobenzoic acid; DAM, 2,4,6-trimethyl-1,3-phenylenediamine; DAPI, diaminophenylindane; DMA, *N,N'*-dimethylacetamide; DMF, *N,N'*-dimethylformamide; DMSO, dimethylsulfoxide; EDA, 1,2-ethylenediamine; GS, gas separation; HF, hollow fiber; IPN, interpenetrating polymer network; NMP, *N*-methylpyrrolidone; MDI, methylene diamine; MET, Membrane Extraction Technology; MW, molecular weight; MWCO, molecular weight cut off; ODP, 4,4'-oxydiphthalic anhydride; OFHD, 2,2,3,3,4,4,5,5-octafluorohexanediol; PAI, poly(amide-imide); PAN, polyacrylonitril; PDMC, 1,3-propanediol monoesterified crosslinkable PI; PDA, 1,3-propanediamine; PEGDA, poly(ethylene glycol) diacrylate; PI, polyimide; PMDA, pyromellitic dianhydride; PEG, poly(ethylene glycol); PPG, poly(propylene glycol); PPGDA, poly(propylene glycol) diamine; PVA, poly(vinyl alcohol); RT, room temperature; SRNF, solvent resistant nanofiltration; *T*_g, glass temperature; TDI, methylphenylene-diamine; TGA, thermogravimetric analysis; THF, tetrahydrofuran; TMPDA, 2,3,5,6-tetramethyl-1,4-phenylenediamine; WAXD, wide-angle X-ray diffraction; XDA, *p*-xylylenediamine.

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1. Introduction

In membrane technology, polyimide (PI) membranes have been used for a long time already and in a wide variety of applications [1]. PI is an excellent polymer to prepare membranes because of its outstanding heat resistance and good mechanical strength, as well as its chemical resistance to many solvents [1]. More recently, the concept of crosslinking PI membranes has become very popular. Crosslinked PI membranes were initially developed for gas separation to reduce the phenomenon of plasticization [2–7] but have since been successfully applied in pervaporation [8–11] and have become very important for solvent resistant nanofiltration processes where the complete range of organic solvents can now be treated with these crosslinked polyimides [12–18]. A variety of crosslinking methods, both physical and chemical, have been described in literature, aiming at different applications. For most of these methods, many different reaction conditions and a large range of crosslinking chemicals have been reported. This review summarizes the extensive literature on crosslinking methods for PI membranes. It focuses on the actual chemistry of crosslinking reactions and the effects on membrane performance. Also, a listing is provided of the main applications for which these membranes have been used. Since it is well known that thermal annealing may further affect the crosslinked membranes and in some cases enhance their performance [10,19–21], this concept is also explored. Some rather new and unexplored crosslinking methods are briefly discussed as well.

2. Polyimide: the material

2.1. Properties of polyimides

PIs, characterized by the presence of the imide group (see Fig. 1) in the polymer backbone, cover a whole range of high performance materials with applications in highly

technical fields, from aerospace to microelectronics [22]. They can be applied as films, fibers, moulding powders, coatings and composites. The major advantage of PIs is their outstanding resistance to heat [22,23], but most PIs also exhibit good mechanical strength as well as a relatively high resistance to chemical solvents.

The structure of the PI backbone determines the heat resistance as well as the chemical resistance of the PI. The rigidity, originating from the combination of the imide structure with an aromatic structure results in polymers with a very high glass transition temperature (T_g). Moreover, the aromaticity of the aromatic groups and the electron-deficiency of the imide bonds guarantees a good oxidative stability [22]. However, the strong heat resistance of PIs also causes processing difficulties. To solve this, the backbone can be ‘diluted’ with other, less stable structures. The polymer then loses some heat resistance but becomes more soluble and processable [24]. The glass temperature of PIs ranges from 280 to 400 °C. Imide bonds are also resistant to commonly used solvents and oils.

With respect to the membrane separations that will be discussed below, the polymer properties that matters most are those that influence the solubility and the diffusivity of the permeating compounds through the crosslinked polyimide, since transport through dense membranes in gas separation, pervaporation and to a large extent also in solvent resistant nanofiltration, generally obeys the solution-diffusion transport model [25–27]. The solution

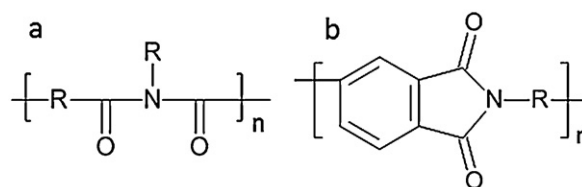


Fig. 1. General structure of (a) a linear PI and (b) a heterocyclic aromatic PI.

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