



## TRENDS IN THE SYNTHESIS OF POLYIMIDES AND THEIR NANOCOMPOSITES

Cite this: *INEOS OPEN*,  
2021, 4 (3), 78–89  
DOI: 10.32931/io2114r

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Received 22 July 2021,  
Accepted 20 September 2021

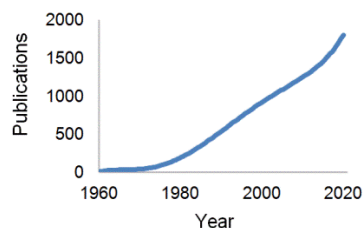
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### Abstract

This review presents the current achievements in the ecologically friendly synthesis of polyimides. The progress in the formulation and applications of different nanocomposites based on polyimides are highlighted. The enormous potential of the polymers and composites under consideration in environmentally benign synthesis and various application fields is shown.

**Key words:** polyimides, nanocomposites, synthesis and application, carbon and inorganic nanoparticles, metal-organic frameworks.

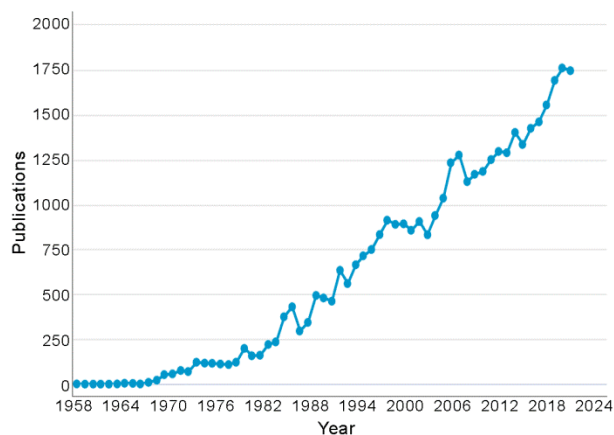


## 1. Introduction

The rapid growth of publications devoted to the synthesis, properties, and different applications of polyimides (PIs) reflects the increasing interest in this class of highly heat-resistant condensation polymers (Fig. 1).

The most popular method for obtaining PIs is a two-step approach [1] that includes the synthesis of a poly(amic acid) precursor by the reaction of a tetracarboxylic acid (TCA) dianhydride with a diamine in a polar solvent at a low temperature at the first step. Then, this precursor is converted to the corresponding PI by intramolecular thermal or chemical imidization.

The second most important approach to PIs is a one-step high-temperature polycyclocondensation of TCA dianhydrides with diamines in a high-boiling organic solvent (*m*-cresol, nitrobenzene, sulfolane, NMP, *etc.*) in the presence of a catalyst: pyridine, isoquinoline, or benzoic acid [2].



**Figure 1.** Publications on polyimides from 1958 till 2020 according to the Scopus database.

Several alternative synthetic approaches to PIs are also known; for example, a convenient and practically valuable reaction of diisocyanates with TCA dianhydrides, which is used, in particular, for the production of PI foams [3–5]. Yet another approach is the Mitsunobu reaction [6, 7] that consists in the interaction of diimides with dihydroxyalkylamines at room temperature and provides direct synthesis of PIs. Liou *et al.* reported [8] a synthetic route based on the reaction of diamines with dithioanhydrides at 140 °C. There are also other methods for obtaining PIs: for example, transition metal-catalyzed reactions of halogenated compounds and diamines or diboronic acid derivatives [9–11], the nucleophilic substitution of dinitro-*bis*-imido-substituted monomers with bisphenols [12], and the use of silylated diamines [13] or mixed diesters/dichlorides of tetracarboxylic acids as monomers. Each of the mentioned methods has its own advantages and drawbacks.

The pollution control promotes a continuous search for new methods for the synthesis of polymers, in particular, PIs that would conform to the green chemistry principles. The last twenty years have witnessed considerable interest in the synthesis of PIs in the new, more ecologically friendly reaction media: melts of organic acids, ionic liquids (ILs) [14–16], and supercritical CO<sub>2</sub> (*sc*-CO<sub>2</sub>) [17, 18]. Of great promise is the hydrothermal and microwave-assisted synthesis of PIs. The following sections describe in detail each of these methods.

## 2. Ecologically friendly synthesis of polyimides

### 2.1. Synthesis of polyimides in *sc*-CO<sub>2</sub>

Non-explosive, nonflammable, and nontoxic *sc*-CO<sub>2</sub> has a high level of ecological safety, in particular, as a medium for the synthesis of PIs. Easy removal of *sc*-CO<sub>2</sub> after the reaction completion, which eliminates the problem of a residual solvent, and its relatively low cost offer huge potential for both

fundamental research and industrial application [19]. The discovered catalytic activity of *sc*-CO<sub>2</sub> in the presence of a trace amount of water facilitates the acylation of an amine; this accelerates the formation of the polyamidoacid and its following cyclization into the PI, which makes this medium even more versatile [17].

The behavior of PIs in the presence of *sc*-CO<sub>2</sub> strongly depends on the polymer structure, its physico-chemical properties ( $T_g$ , crystallinity), the purity of *sc*-CO<sub>2</sub>, as well as the system temperature and pressure. It is believed that the solubility, a key factor in the synthesis of polymers, and inhomogeneity of a reaction medium hamper the production of polymers with high molecular weights [20]. However, several methods have been suggested to overcome the problem of inhomogeneity [17, 21]. It was noted that the latter does not play a crucial role in hampering the production of high-molecular PIs (yield 90%,  $\eta_{inh} = 0.56$  dL/g). Among the important factors, the authors highlight an increase in the reaction duration and temperature, the pressure of *sc*-CO<sub>2</sub>, synthesis in the presence of a trace amount of water, and the application of monomers with fluorine- and/or siloxane-containing moieties.

Besides the advantages of *sc*-CO<sub>2</sub> for the environment, it can form nanoscale pores in a polymer upon removal from the reactor. This enables considerable modification of the microstructures of PIs and materials on their base and, thereby, offers new opportunities for the production of heat-resistant foams, aerogels, catalysts, separation membranes, and so on [22].

Although the cost of the equipment for working under supercritical conditions is relatively high and the current technologies for the synthesis of PIs in supercritical fluids are far from the replacement of traditional methodologies, the results obtained testify to the great potential of this field. The appearance of new materials and technical solutions make the methods that can be realized at elevated temperatures and pressures more available and economically reasonable.

## 2.2. Synthesis of polyimides in ILs

One of the application fields of ILs is the chemistry of high-molecular compounds. The catalytic activity caused by the ionic nature of ILs provides an increase in the rate of polycondensation and a reduction in its temperature [23]. Moreover, ILs possess low vapor pressure and do not mix with many organic solvents [16]. ILs facilitate the isolation of the target product in a quantitative yield and with the inherent viscosity ( $\eta_{inh}$ ) ranging from 0.1 to 2.2 dL/g and enable solvent regeneration for reuse [24].

Firstly, ILs were suggested for the synthesis of classic PIs by the two-step methodology *via* polycondensation of industrial diamines with TCA dianhydrides. It was found that the good solubilizing ability and catalytic properties of ILs ensure their application for the polycondensation of exotic and low-reactive building blocks [25, 26].

Nowadays, ILs are used not only as a new class of ecologically friendly solvents but also as monomers for polymer analogs of ionic liquids (PILs) [27]. A small content of ionic moieties in the PI structure affects the thermomechanical or gas separation properties of the resulting films and membranes. The synthesis of PILs is usually accomplished by the

polycondensation of the classic building blocks with ionic monomers (most frequently, diamines) or through the modification of non-ionic PIs [28]. Compared to the neutral precursors, PILs display improved solubility, enhanced hydrolytic stability, higher gas permeability, and controlled hydrophobicity [29].

Over the last two decades, ILs greatly shaped polymer chemistry and materials science. The widespread application of this new class of environmentally benign solvents can facilitate a reduction in the use of polluting conventional organic solvents and the synthesis of macromolecules with specific operational characteristics.

## 2.3. Synthesis of polyimides in acid melts

The preparation of soluble thermoplastic PIs of linear and branched structures in the benzoic acid (BA) melt was developed over 20 years ago [30]. The synthesis of PIs utilizes diamines and aromatic TCAs or their derivatives (dianhydrides, diesters). The method conforms to the green chemistry principles since it combines a nontoxic medium with a simple isolation procedure of the target polymer. The advantages also include a homogeneous mode of the reaction independent of the diamine basicity and mild synthesis conditions (140 °C, 1–2 h) [31, 32]. However, there are also some limitations. Thus, BA readily sublimates at 100 °C, which requires the hermetically sealed conditions for the reactions.

Later, the method was suggested that is based on a one-step synthesis of PIs in the salicylic acid melt [33]. Hasanain *et al.* supposed that this medium has higher acidity ( $pK_a = 2.97$ ), lower volatility, and excellent solubilizing ability compared to conventional *m*-cresol, *p*-chlorophenol, and BA ( $pK_a = 4.19$ ). The yields of PIs varied from 67 to 99%, and the values of  $\eta_{inh}$  ranged within 0.5–1.0 dL/g.

This method can be recommended for the production of polyimide constructional thermoplastics, binding agents for polymer composite materials, electroinsulating films, and gas separation membranes.

## 2.4. Solvothermal synthesis of polyimides

About 10 years ago, an unusual synthesis of PI was described that involved the application of water (or ethanol, isopropanol, and glycerol) as a solvent at high temperatures (180–200 °C) and pressures (up to 16.7 bar) [34–36]. The reactions of tetracarboxylic acids with diamines afforded nylon-type salts at the first step [36]. Further heating above 180 °C converted the resulting salts to highly crystalline PIs in the presence of water. It was noted that the selective fractionation can afford PIs with the weight-average molecular weights ( $M_w$ ) from 4400 to 50000. Due to its resemblance to the hydrothermal conditions of silicate formation in the earth crust, this method for the PI synthesis was called hydrothermal polycondensation [37].

As well as the above-mentioned methods, this synthetic approach can be considered ecologically friendly owing to the absence of common solvents and catalysts. The reaction proceeds as a classic polycondensation and affords, firstly, semicrystalline oligomeric PIs from amorphous oligoimides in a low-temperature hydrothermal mode (100–130 °C), which coexist with zwitterion dimers. At 200 °C, the morphological

ordering increases, and the formation of highly crystalline PIs is observed [35]. Experimentally it was shown that the prevention of a competitive solid-state polycondensation creates impressing morphological uniformity and reduces the content of the amorphous PI [38]. The production of highly crystalline polymers was confirmed by the powder XRD and SEM studies. The beginning of decomposition of these PIs in a nitrogen atmosphere was observed at approximately 600 °C [39].

Despite the fact that the developed hydrothermal method cannot replace the conventional methods for the synthesis of PIs due to its complicated equipment implementation, it opens new prospects for the production of polymers and materials which are difficult to obtain by the traditional techniques. For example, the covalent binding of a porphyrin unit with the PI macromolecules allowed Cui *et al.* [40] to enhance the absorption of visible light in 10 times compared to that of the PI lacking the porphyrin moiety. The photocatalysts based on copper oxide nanocomposites dispersed on the PI surface allowed for a 32-fold increase in the rate of H<sub>2</sub> formation [41]. The resulting microporous amorphous PIs [42] can separate CO<sub>2</sub> from N<sub>2</sub>, which can become an interesting addition to classic microporous materials.

## 2.5. Synthesis of polyimides under microwave irradiation

In recent years, the ecologically friendly and rapid synthesis of PIs under microwave irradiation has been developed. This method allows one to perform the polycondensation over short periods of time and avoid energy and time-consuming isolation and purification of PIs.

The films from the PIs obtained as a result of microwave heating were shown to possess higher tensile strength, elastic modulus [43], and optical transparency than those from the polymers obtained by thermal cyclization, which enables their use in OLED fabrication [44]. Insignificant drawbacks of the microwave-assisted synthesis include a necessity to dissolve PIs for the production of films and membranes. Furthermore, PIs can partially destruct due to continuous or too intense irradiation [45].

Son *et al.* synthesized PIs based on phenylenediamine with branched highly phenylated moieties which provide enhanced solubility and processability [46]. The aromatic PIs obtained in 30 min featured  $M_w = 30000 \div 59300$  and polydispersity ranging from 1.6 to 1.9. At the same time, they exhibited high optical transparency and excellent solubility in chloroform, tetrahydrofuran, toluene, and cyclohexanone. The materials based on these PIs can find application in fuel cells, gas separation membranes, coatings, and composite materials.

Zhuo *et al.* [47] suggested the synthesis of PI by the solvothermal method in ethanol using microwave heating. At the initial step, the intermediate tetracarboxylic acid diesters and soluble ammonium salts were obtained. The microwave impact on the salts resulted in crystalline PIs.

This method has a number of highly valuable and solicited advantages as an ecologically friendly and economical approach to PIs. Despite the technological problems and limitations, it has a great potential for further development.

## 3. Synthesis of nanocomposites based on polyimides

Over the last two decades, significant advances have been made in the field of synthesis and investigation of the physico-chemical properties and applications of (nano)composites based on PIs. Among them of particular interest are the molecular composites of a polymerization-polycondensation type, nanocomposites with carbon nanotubes, graphene, inorganic fillers, and metal-organic frameworks. This review highlights the main aspects of their production and possible application fields.

### 3.1. Molecular composites of a polymerization-polycondensation type

The molecular composites (MCs) are polymer-polymer systems in which the rigid-chain macromolecules are dispersed at the molecular level over a matrix of flexible polymers, where the rigid-chain molecules act as reinforcing agents [48]. The MC components must satisfy the following requirements: the reinforcing agent in the form of the long rigid-chain macromolecules is uniformly distributed in the flexible matrix; the rigid-chain polymer and the flexible matrix must be thermodynamically compatible and mix at the molecular level without phase separation [48, 49]. The peculiarities of the formation of molecular composites, their diversity and properties are described in detail in several reviews [48, 50, 51].

The establishment and development of the MC concept predetermined an interest in the construction of PIs that would differ by the enhanced thermodynamic rigidity along with the high strength of fibers and films, heat resistance, and, at the same time, good solubility in organic solvents. Such a combination of the properties distinguishes PIs from many other rigid-chain polymers as promising reinforcing agents [51].

The mechanical blending of different polymers at the molecular level is considered to be one of the most important methods for the development of new polymer compositions with the required properties [52, 53]. The simplicity, reproducibility, and possibility of wide variation of the composite properties are the most important advantages of these systems. Furthermore, a synergistic effect allows for obtaining materials with the properties that are not characteristic of any of the starting components [54–56].

The mechanical blends of PIs with different flexible-chain polyimides [57–59], polyamides [60–64], polysulfones [65–67], polyester imides [68], polyphosphazenes, and other polymers [53, 69] found wide application in the production of membranes for gas separation, purification of organic solvents, fuel cells, *etc.* [53, 56, 65, 70–72]. Mixing the polymers at the molecular level was accomplished in a solvent (NMP, DMA, DMF, and THF) at the component concentrations below the critical ones, *i.e.*, in the absence of phase separation. The resulting membranes appeared to be highly efficient for the separation of H<sub>2</sub>/CO<sub>2</sub> [58, 64, 65], CO<sub>2</sub>/CH<sub>4</sub>, O<sub>2</sub>/N<sub>2</sub>, CO<sub>2</sub>/N<sub>2</sub>, He/N<sub>2</sub> [63, 64], H<sub>2</sub>/CH<sub>4</sub>, H<sub>2</sub>/O<sub>2</sub>, H<sub>2</sub>/N<sub>2</sub> [58, 64], and other gases [53, 69]. Furthermore, the authors noted the flexibility and thermal stability of the composite membranes.

The method for obtaining membranes by pyrolysis of mixtures of semiaromatic polyetherimides with aromatic imides was described [68]. The high gas separation characteristics, resistance to plasticizing, as well as remarkable mechanical properties of the films were caused by the partial cross-linking of the macromolecules as a result of thermal processing.

The potential of membranes based on mixtures of sulfonated PIs and polyphosphazenes for vanadium batteries was demonstrated [70]. The membranes were obtained by the classical method from a solution of PI with 1 wt % of poly-*bis*-(4-methylphenoxy)phosphazene in NMP. The formed membranes demonstrated resistance to oxidation and hydrolysis, long service lives, as well as high proton conductivity and low permeability for vanadium ions.

The hybrid films based on PI–polybenzoxazine obtained by the thermal curing of a mixture of benzoxazine with different polyimides [73–78], polyamidoacids [73–77], or polyimide–siloxanes [73, 74, 76, 77, 79, 80] are described. The composites have a single glass transition temperature and are characterized by high temperature resistance and mechanical properties, which makes them promising for microelectronics [73, 76].

Despite the attractiveness of the mixed MCs, an essential drawback that restricts their design and application is the incompatibility of rigid rod-like macromolecules with a flexible matrix. The materials obtained from partially or incompatible components often have low thermal and mechanical stability [53, 69, 72].

It seems very promising to synthesize block and graft copolymers in which the rigid and flexible fragments are connected by stronger covalent bonds. This not only allows for increasing the degree of compatibility of the components owing to the strong intermolecular interactions but also creates a possibility for fine-tuning their morphology and properties [51, 69].

The block and graft copolymers are obtained, as a rule, by terminal groups or substituents in the PI main chain, which acts as a macromolecular activator or polymerization initiator. The terminal groups can be both the PI own functionalities (amino or carboxy groups) or specially introduced moieties.

The atom-transfer radical polymerization of the monomers such as MMA [81–83], methacryl-containing polyhedral oligomeric silsesquioxane [84–86], 2-methyl-2-adamantyl methacrylate [87, 88], and 2-hydroxyethyl methacrylate [89] afforded a series of ABA block copolymers. The selective decomposition of the polyacrylate blocks that proceeds prior to the beginning of PI decomposition afforded films promising for microelectronics ( $k = 2.36$ ) that featured the pore sizes of 50–100 nm [81] and gas separation/sorption membranes [87, 88, 90]. The films from the PMMA–block–PI–block–PMMA copolymers exhibit high gas barrier [82] and hydrophobic properties [83] as well as low refractive indices [82]. The presence of the polyhedral oligomeric silsesquioxane moieties leads to enhanced hydrophobicity and improved gas separation properties compared to the unmodified PIs [84–86].

High-impact ABA-type copolymers were obtained by the anionic activated polymerization of  $\epsilon$ -caprolactam in the presence of PI with grafted terminal, usually acylactam [49, 51, 91] and less often other polymerization-activating groups [92, 93]. An alternative approach is proposed in which the PI with side phenyl ether groups were used for polycaprolactam grafting

[93, 94]. With a PI content of only 5 wt %, the copolymers are superior to the corresponding homo-polycaprolactam in the thermal and moisture resistance as well as in mechanical characteristics in 2–3 times [51, 91–94].

The synthesis of graft copolymers based on the PIs bearing side functional groups that act as polymerization initiators for the following vinyl monomers was described: styrene [95–97] and different (meth)acrylates [96, 98–104]. There was a significant increase in the rigidity of the main polymer chains of PI–graft–polystyrene (by an order of magnitude) compared to the initial PI [97] at the maintained mechanical strength [95, 104]. The prospects of application of these composite systems in membrane technologies [99, 105], microelectronics [95, 104], measuring and control systems [96, 103] have been demonstrated. The peculiarities of the interaction of rare-earth and transition metal ions with some of the resulting copolymers were studied [106]; the latter can find application as nanoreactors or nanocontainers [98, 107].

A principal possibility of the application of PIs without preliminary functionalization in the anionic polymerization of  $\epsilon$ -caprolactam was demonstrated [108]. Under the action of a polymerization catalyst, MgBr- $\epsilon$ -caprolactam, the PI imide ring opens to yield a graft comb-shape copolymer. The resulting copolymers are characterized by high mechanical strength, heat resistance, and improved tribological properties; the impact strength of the samples is several times higher than that of not only homo-polycaprolactam but also the corresponding block copolymers [108–110].

The free-radical *in situ* (co)polymerization of MMA [111–113], ethyl acrylate [114, 115], fluoro(meth)acrylates [116, 117], MMA with bifunctional comonomers [118], styrene [119], styrene with MMA [119], *N*-vinyl-2-pyrrolidone [120, 121] in the presence of PI (4–85 wt %) afforded the molecular composites of different formulations. The mechanism of copolymer formation that consists in the interaction of free radicals of the reaction system with macromolecules of PI has been established by DSC and EPR [111, 114]. The products of thermal or photopolymerization are the copolymers that strongly differ in the properties from the corresponding homopolymers and their mixtures [111–121].

In conclusion, it should be noted that the molecular composites based on a combination of two or more polymers, including polymer blends and different copolymers, can be very promising for the production of films, membranes, constructional plastics that find application in different fields. The technology of fabrication of multicomponent polymeric systems as a modern method allows one to obtain a range of materials with versatile characteristics.

### 3.2. Nanocomposites with carbon nanotubes

Carbon nanotubes (CNTs) can be conditionally divided into single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). Under certain conditions, they can acquire the properties which are usually associated with traditional molecular particles, such as stability of dispersions in organic solvents, chemical transformations based on solutions, chromatography, and spectroscopy [122].

The phase separation and high propensity for agglomeration are the main problems of effective inclusion of CNTs into PI

matrices. The system dispersity can be improved by functionalization using surfactants or ultrasonic processing. Among the mentioned approaches, the most efficient ones are covalent [123] or noncovalent [124] functionalization of CNTs.

Huang *et al.* [125] demonstrated an efficient method for the covalent functionalization of carboxylated nanotubes upon simultaneous ultrasonic treatment. The resulting SWCNT/PI films displayed 10 times higher electroconductivity at the low SWCNT loading (0.1 vol %). These composition films remained optically transparent. The mechanical properties and thermal stability were also improved owing to the introduction of the CNTs into the PI matrix. For better compatibility with CNTs, the PIs with terminal amino groups [126, 127], side hydroxy [128] or cyano [129] groups were synthesized. The stabilization of the CNT dispersion occurs owing to the covalent or donor-acceptor interaction.

The addition of insignificant amounts of CNTs to PIs reduces their electrical resistance and eliminates the problem of accumulation of a static charge in polymeric materials. It was established that the conductivity of the composites follows a percolation power law with a relatively low percolation threshold (0.05 vol % of SWCNT). Depending on the SWCNT concentration, the composite conductivity can be enhanced by several orders of magnitude [130, 131].

At the good distribution of CNTs and their affinity to the matrix, they act a role of reinforcing agents. It was shown that the addition of 1 wt % of MWCNTs leads to a 250% increase in the nanocomposite elasticity, whereas the tensile strength is comparable to that of the matrix [131]. The strengthening is associated with a strong interphase interaction between the MWCNT and PI matrix, which facilitates the stress transfer from the polymer to the CNTs. This is evidenced by the SEM images of the fracture surfaces [132] that depict strong interphase adhesion (the presence of a multitude of broken but strongly held CNTs in the polymer matrix).

An example of noncovalent functionalization was reported [124, 133] where the carboxylated SWCNTs act as crystallization centers for the amorphous PI matrix. The low SWCNT concentrations (0.1–1.2 vol %) led to an increase in the heat resistance of the composite by 100 °C and a double increase in its impact strength compared to the initial polymer. It was established that the main PI chain must be flexible in order to allow the aromatic rings to adopt an energetically favorable conformation, being located tangentially to the surface of the SWCNT and thereby increasing the  $\pi$ - $\pi$  interactions [134].

Zhou *et al.* [135] showed that photoluminescence is an efficient method for evaluating the distribution of functionalized SWCNTs and can be used to study the CNT dispersion in polymer nanocomposites since the luminescence emission is sensitive to the degree of agglomeration of nanotubes into beams and modification of the surface.

The PI nanocomposites with CNTs will play an important role in a large variety of applications, for example, in materials for protection from static electricity, radiofrequency devices, flexible displays, and other modern gadgets [136].

### 3.3. Nanocomposites with graphene

A large number of works are devoted to the PI nanocomposites with graphene. The relative availability and

unique properties of graphene (high conductivity, thermal conductivity, strength, and hydrophobicity) attract considerable interest of not only researchers but also industry, including large companies such as Samsung, IBM, *etc.* The original graphene, as well as another allotropic modification of carbon—CNT, due to its tendency to agglomeration, is not suitable as a nanofiller. Therefore, modified graphenes, for example, graphene oxide (GO), fluorinated graphene, or graphene functionalized with different compounds are used [137].

It was noted that the composite PI films obtained *in situ* from solutions with partially reduced GO have considerably enhanced thermal and electroconductivity [138, 139]. Thus, for example, the addition of 3.0 wt % of GO to PI allows one to achieve the electroconductivity of  $5.38 \cdot 10^{-5}$  S/m and the thermal conductivity of  $0.10 \text{ Wm}^{-1}\text{K}^{-1}$  [138]. This was accompanied by a 75% increase in the tensile strength. The addition of 20 wt % of GO to PI allowed for reducing the resistivity of the films almost by 9 orders of magnitude [139]. The ultimate tensile strength of this composite film decreased from 113 MPa to 99 MPa, and the elastic modulus increased to 4.4 GPa vs. 2.3 GPa for the initial PI film. An increase in thermal and electroconductivity offers opportunities for the production of flexible heating elements based on the PI/GO nanocomposites. The electrical resistance of the resulting flexible heater after 9000 bending test cycles reduced less than by 1% [140]. The test for a heating rate to 100 °C with a sheet featuring the surface area of 100 mm<sup>2</sup> demonstrated that, at the voltage of 32 V, the required temperature can be reached in 16 s.

Recently, new highly promising cathode materials for Li- and Na-ion batteries based on the PI and graphene nanocomposites were suggested [141, 142]. These materials feature high mechanical strength without the addition of any other binding or conducting carbon, high reversible capacity ( $240 \text{ mA}\cdot\text{h}\cdot\text{g}^{-1}$  and  $40 \text{ mA}\cdot\text{g}^{-1}$ ), excellent performance ( $102 \text{ mA}\cdot\text{h}\cdot\text{g}^{-1}$  at  $4000 \text{ mA}\cdot\text{g}^{-1}$ ), and remarkable cyclic stability (capacity retention of 81.2% after 600 cycles at  $100 \text{ mA}\cdot\text{g}^{-1}$ ). A flexible electrode provides for ultra-high accumulation capacity equal to  $213 \text{ mA}\cdot\text{h}\cdot\text{g}^{-1}$  at  $50 \text{ mA}\cdot\text{g}^{-1}$  and capacity of 80.4% after 1000 cycles at  $1000 \text{ mA}\cdot\text{g}^{-1}$ . The high characteristics of the cathode can be explained by rapid redox processes, rapid electron transfer, and advantages of ionic conductivity owing to an increase in the surface area in the nanocomposites.

The PI/graphene nanocomposites are promising also for friction materials. Thus, Huang *et al.* [143] accomplished the polycondensation of 4,4'-(4,4'-isopropylidenediphenoxy)-bis-phthalic acid anhydride and 4,4'-oxydianiline in the presence of graphene modified with aminophenyl groups. The resulting nanocomposite showed a 20-fold increase in the wear resistance and a reduction in the friction coefficient by 12%. Wu *et al.* [144, 145] modified graphene with L-phenylalanine in the presence of polyphosphoric acid or by the reaction of 1,3-dipolar cycloaddition of N-methylglycine and 3,4-dihydroxybenzaldehyde. To obtain a wear-resistant nanocomposite, the modified graphene (up to 2.0 wt %) was mixed with PAA and subjected to thermal cyclization on a substrate. It is noted that with the improvement of the tribological properties of composites, their thermal stability, electrical conductivity and mechanical properties, including the tensile strength, increased. The authors explain the improvement

of the properties by strong adhesion interaction at the interphase boundary and production of a uniform oriented dispersion.

Besides the tribological advantages, the PI/graphene nanocomposites exhibit also good hydrophobic and gas separation properties that provide anti-corrosion protection. The homogeneous distribution and strong interphase interaction between the aminophenyl-functionalized GO and PI matrix afforded nanocomposite films with a reinforcing effect and barrier properties from oxygen penetration that exceed the analogous PI by 99% [146]. These thin nanocomposite films have great potential for application as substrates in modern flexible electronic devices [147]. To use in microelectronics and aerospace industry, these materials must exhibit, besides the mentioned mechanical and hydrophobic properties, low permittivity. It was found [148, 149] that the addition of 0.5 wt % of fluorinated graphene to PI affords a composite with excellent mechanical properties of the films, low dielectric permeability equal to 2.48, and specific conductivity no less than  $10^{-14}$  S/m. Due to the development of high-speed communication that operates through the 5G standard, polyimides with a dielectric permeability below 3 will be required for wireless communication antennas [150].

The application scope of the PI/graphene composites is not limited to classic devices. A highly sensitive pressure sensor was obtained from the PI foam and graphene using an ecologically friendly technique [151]. The piezosensors of different shapes and thicknesses were obtained by wetting the PI foam in dispersed GO followed by the reduction with ascorbic acid ( $\gamma$ -2,3-dehydro-l-gulonolactone) and annealing at 80 °C for 6 h. The samples obtained displayed high sensitivity of  $\sim 0.36$  kPa<sup>-1</sup>, which enables their use as heartbeat, hinge mobility, and air flow measuring sensors.

Feng *et al.* [152] described a successful production of the PI/GO pervaporation membrane that is able to trap about 99.9% of ions from sea water. Its permeability was 36.1 kg·m<sup>-2</sup>·h<sup>-1</sup> at 90 °C and 0.1 MPa. The productivity of desalination units based on these hybrid membranes remained unchanged for 120 h at 75 °C. The results obtained are positive indicators of a potential candidate for desalination of sea and brackish water.

### 3.4. Nanocomposites with inorganic fillers

In recent years, increasing attention has been given to the creation of multifunctional hybrid organic-inorganic materials that include both organic and inorganic components [153, 154]. The variation of the component ratio opens the way to the creation of composite materials with the required strength characteristics, elastic modulus, and heat resistance, as well as compositions with the specific magnetic, dielectric, optoelectronic, radar-absorbing, and other properties. These composite materials are extensively used in opto-, photo- [155–158], and microelectronics [159, 160], aerospace industry [161, 162], membrane technologies [163–166], and other fields [150, 167–169].

Among a great variety of inorganic fillers, of particular attention are layered and skeleton silicates [150, 153, 170–172], metal oxides [153, 160], polyhedral oligomeric silsesquioxanes [153, 173–177], and others [178].

Layered (sheet) silicates such as clay, silica, and montmorillonite, which have, on the one hand, nanosized

layered particles and, on the other hand, available raw materials, represent attractive components for modern composite materials [153]. The clay layers with a thickness of about 1 nm are connected by ionic or Van der Waals forces. The problem of nanoparticle aggregation into large agglomerates, which is characteristic of many nanofillers, can be solved by the high propensity of layered silicates for swelling, which facilitates intercalation of the monomer or polymer molecules into their interlayer space followed by exfoliation of the filler particles into monolayers [171, 179]. This results in the uniform distribution of the nanoparticles over the polymer matrix.

Among the possible methods for producing nanocomposites from PIs and layered silicates, the mechanical blending of the polymer melt with silicate and *in situ* polymerization are used. However, the method of forming composites from the PI solutions containing nanoparticles has become the most widespread one [171].

The casting of the PI solution with the silica content up to 37 wt % afforded flexible optically transparent films with high thermal stability (above 400 °C) [167]. It was revealed that filling above 40 wt % leads to phase separation, turbidity of the films, and loss of mechanical strength.

Yu *et al.* [180] suggested the method for the production of pearly composite films by gravitation precipitation of the preliminarily exfoliated montmorillonite (50 wt %) in the PI solution. The prominent mechanical and vapor-isolating properties, in particular, under conditions of enhanced humidity make this composite a potential candidate for electronic encapsulation with the enhanced requirements to threshold properties, such as fixed solar cells and OLEDs.

The filling of a fluorinated polyimide with terminal carboxy groups up to 50 wt % of silica afforded flexible films with the optical transparency both in the UV and visible regions. It was assumed that the terminal carboxy groups undergo condensation with colloidal silica, thereby, suppressing the particle agglomeration and interphase separation [181].

A similar approach was used to obtain hybrid polyimide composites by the *in situ* polycondensation in the presence of SiO<sub>2</sub> with grafted amino groups [182]. The covalent binding of the filler and PI led to the improvement of strength and elastic modulus (by 25% and 40%, respectively) at the filling levels of 2–10 wt %.

Similar results were obtained in a series of works devoted to the application of different triethoxysilanes as cross-linking agents [164, 166, 183–185]. The comparative studies showed that the PI films with covalently bound silica exceed the films with unmodified SiO<sub>2</sub> in the mechanical properties [183].

A possibility of using graphene oxide as a dispersing agent to improve the dispersibility of montmorillonite in the PI solution was demonstrated [186]. The resulting hybrid materials were used for the production of reinforced composite PI aerogels with high compression modulus (14 MPa), thermal and fire resistance.

Intercalation *in situ* polycondensation in the presence of layered silicates was used to obtain polyimide nanocomposites with the addition of special swelling agents. The swelling agent can more easily penetrate to the interphase space of montmorillonite, thereby, splitting the plates followed by exfoliation of the filler particles under the action of the resulting polymer [171, 179].

This approach was used for the production of hybrid polyimide films with good nonlinear optical properties [157]. The chemical grafting was accomplished using a special trifunctional swelling agent, namely, 1,1,1-*tris*[4-(4-amino-2-trifluoromethylphenoxy)phenyl].

A series of reports, aimed at the production of effective polymeric composite materials, suggested the modification of layered aluminosilicates by replacing hydrated alkali and alkaline earth metal cations in the montmorillonite lattice for organic cations. The introduction of organic cations, most frequently alkyl ammonium ones, leads to an increase in the interplanar distance in the silicate lattice, which allows one to realize an exfoliation effect of layered fillers in the composite material matrix [156, 187–189]. Mansoori *et al.* suggested the substitution of metal cations for aromatic diamines in order to obtain more thermally stable montmorillonite [190]. It was shown that the composition and structure of the organic cation define the mechanism of intercalation and exfoliation degree of silicate to nanoscale plates in the polymer composite [150, 179]. As the concentration of the organic cations increases, the interphase distances also increase and the exfoliation to nanoplates is simplified, what contributes to improving the elastic-strength and barrier properties of the composites [150, 169, 179].

Polyhedral oligomeric silsesquioxanes (POSSs) amount to one of the most promising nanomaterials and attract continuous research interest. The chemical structure of POSSs can be presented by the formula  $(\text{RSiO}_{1.5})_n$  where  $n = 6, 8, 10, \text{etc.}$ , R is an organic radical at an external side of the core and bearing a reactive group (for example, alkyl, alkylene, epoxy unit, acrylate, hydroxy group) or a hydrogen atom [133, 173, 175]. The organic substituents around the inorganic core facilitate better dispersing of POSS in solvents and in a polymer matrix as well as covalent binding of nanoparticles with the polymer macromolecules, which distinguishes them from other fillers. The application of composites bearing POSS is defined, first of all, by their high mechanical properties, fire-resistant and electrical characteristics combined with thermooxidative stability, which is particularly important in fuel cells, microelectronics, and aerospace industry [159, 161, 162, 175–177].

Atar *et al.* [161] solved the problem of the creation of flexible electroconductive films with a high level of protection from atomic oxygen by the infiltration of POSS–PI mixtures into carbon nanotube sheets. The latter represent films with a thickness of  $9 \pm 2 \mu\text{m}$  obtained by the chemical precipitation of nanotubes from vapor on a silicon substrate. They were impregnated with a POSS/PI solution in NMP followed by the thermal removal of the solvent and film formation. The PI matrix gives mechanical stability, strength and flexibility to the composite film, and the continuous network of CNTs provides electroconductivity. The suggested method allows one to avoid agglomeration and degradation of the nanotubes, as it was observed during the formation of films from solutions under conditions of high-power ultrasonic treatment. It is also applicable for the production of films with larger areas. Under the action of atomic oxygen, POSS is oxidized to  $\text{SiO}_2$  and creates a protective network on the film surface. The high efficiency of the resulting composite towards the following aggressive factors that simulate the material presence on the

Earth's orbit was established: atomic oxygen, ionizing radiation, and temperature drop.

Song *et al.* [162] showed a possibility of production of heat-resistant phosphorus-containing PI–POSS nanocomposites by *in situ* polycondensation. The erosion of the composite film after atomic oxygen impact reduced by 51%. The performed investigations revealed the formation of two passivating surface layers of phosphate and/or metaphosphate as well as silicon oxide layer under the action of atomic oxygen.

Huang *et al.* [159] suggested a composite material with the improved mechanical ( $\sigma_r \sim 112 \text{ MPa}$ ) and thermal properties ( $T_{5\%} > 550 \text{ }^\circ\text{C}$ ) as well as low dielectric constant ( $\kappa \leq 2.6$ ). The PI with the covalently bound POSS was obtained by the *in situ* polycondensation in the presence of aminopropylisobutylpolysilsesquioxane followed by the film formation. The resulting material holds great promise for the application in microelectronics in the production of 5G chips and millimeter-wave antennas.

### 3.5. Nanocomposites with metal-organic frameworks

Owing to the unique identity of pores and their number, metal-organic frameworks (MOFs) are attracting increasing attention. In 2020, the journal *Chemical Reviews* published a special issue on MOFs. The published reviews present a variety of computational methods [191–193] and synthetic approaches [191, 194–198] as well as different application fields of MOFs: gas separation [199, 200], heterogeneous catalysis [199, 201], separation and sorption of toxic industrial gases [202], water purification from organic pollutants and salts [203, 204], stimuli-sensitive [196, 199, 205], electro- [206], and proton-conducting materials [207], magnets [208], and other areas [199].

The composites based on PIs and MOFs are promising for different applications fields, the most important of which are membrane technologies. Matrimid 5218 polyimide that possesses intrinsic porosity is one of the most popular polymers for the separation of different gases. To endow the membranes with new gas transporting properties, Matrimid 5218 was modified with the MOFs based on salts of zinc and 2-methylimidazole, ZIF-8 [209] and ZIF-90 [210], copper salts of benzene-1,3,5-tricarboxylic acid, HKUST-1 [211], and nanolayers based on MOFs [212]. In all cases, an increase in the permeability of the mixed membranes towards the following gases was detected:  $\text{H}_2$ ,  $\text{N}_2$ ,  $\text{O}_2$ ,  $\text{CO}_2$ , and  $\text{CH}_4$  [209–212]. It was shown that mixing the polymer with ZIF-90 [210] as well as copper salts of terephthalic acid CuBDC [212] leads to doubling of the membrane permeability towards  $\text{CO}_2$  with the maintained initial selectivity of  $\text{CO}_2/\text{CH}_4$  separation.

The addition of hydrophilic HKUST-1 (40 wt %) enabled the production of mixed membranes with the improved separation characteristics for water/ethanol: the pervaporation of water increased twofold without an essential change in the separation coefficient ( $>200$ ) [211]. These membranes were successfully used for shifting an equilibrium during the esterification of acetic acid with ethanol.

Homo- and copolyimides based on 2,2-*bis*-(3,4-dicarboxyphenyl)hexafluoropropane dianhydride (6FDA) and diaminesethylene (DAM) produced by Akron Polymer

Systems (USA) are the newest promising polymer matrices for membrane technologies. The doping of 6FDA-DAM polyimide with ZIF-90 [210] and ZIF-94 [213] afforded a considerable increase in the gas transporting characteristics of the membranes towards the following gas pairs: CO<sub>2</sub>/CH<sub>4</sub> [210] and CO<sub>2</sub>/N<sub>2</sub> [213]. The highest growth of CO<sub>2</sub> permeability (~200%) and unchanged selectivity of CO<sub>2</sub>/N<sub>2</sub> separation (~22) were observed in the case of the membranes with 40 wt % of ZIF-94 [213].

The membranes based on 6FDA-DAM polyimide and ZIF-8 also displayed considerably improved characteristics for the separation of a propylene/propane mixture [214]. An increase in the permeability towards propylene to 56 Barrer (258%) and ideal selectivity of the separation of a propylene/propane mixture up to 31 (150%) were detected for the membrane bearing 48 wt % of ZIF-8 [214].

As well as other nanoparticles that are introduced into polymer matrices, MOFs can also exhibit insufficient compatibility with a polymer, which leads to nonuniformity and defectiveness of the resulting membranes. One of the solutions to this problem is the functionalization of a polymer matrix with the groups that can efficiently interact with the MOF. The excellent compatibility with ZIF-8 was reached upon application of the hydroxy-functionalized microporous PI [215]. The high filling degree and uniformity of distribution of nanoparticles over the matrix were caused by the formation of hydrogen bonds between the components, which was confirmed by the IR and photoelectron spectroscopic studies. The resulting mixed membranes demonstrated unprecedented separation characteristics for a propylene/propane mixture and outstanding resistance to plasticization [215].

An alternative approach consists in the modification of the preformed hollow-fiber membrane with different MOFs. Thus, the modification of the internal surface of industrial membrane P84 with ZIF-93 nanoparticles enabled an increase in the separation selectivity of H<sub>2</sub>/CH<sub>4</sub> and CO<sub>2</sub>/CH<sub>4</sub> pairs to 97 and 17, respectively [216].

In order to improve the compatibility between a matrix and filler, the additional functionalization can be accomplished not only for the polymer but also for the introduced MOFs. Xie *et al.* [217] used both of these approaches simultaneously. Firstly, in order to activate inert PI fibers, their surface was treated with dopamine and, then, using supramolecular assembly, NH<sub>2</sub>-MIL-101 salts functionalized with amino groups were applied to the fibers. The resulting composite fibers were used for the production of high-temperature filters to trap harmful dust [217].

Obviously, the PI–MOF composite membranes have great scientific and commercial potential. The structure of the PI in use, composition formulation and its properties predetermine the potential application of the membrane. Furthermore, the PI–MOF composites can be used in the production of coatings, sensors, and so on.

#### 4. Conclusions

The presented literature review showed that the last two decades have seen considerably increased attention in the synthesis of PIs and composites on their base. Although it is early to talk about the replacement of conventional synthetic approaches to PIs, of note are significant advances in the more

ecologically friendly methods for the synthesis of polyimides. Ionic liquids, supercritical CO<sub>2</sub>, microwave irradiation, and hydrothermal synthesis often lead to the polymers and their composites featuring specific properties. The materials based on PIs and their various composites can exceed the earlier obtained analogs in the characteristics and find applications as gas, proton, ion-exchange, and liquid membranes, electronic devices, aerogels, foams, sensors, *etc.* All this demonstrates a huge potential of PIs for the application in manufacturing devices of different assignments.

#### Acknowledgements

This work was supported by the Russian Foundation for Basic Research, project no. 18-29-17035 mk, and the Ministry of Science and Higher Education of the Russian Federation.

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#### References

1. *Advanced Polyimide Materials: Synthesis, Characterization, and Applications*, S.-Y. Yang (Ed.), Elsevier, Oxford, **2018**.
2. V. S. Ivanov, A. S. Yegorov, G. R. Allakhverdov, V. V. Men'shikov, *Oriental J. Chem.*, **2018**, *34*, 255–264. DOI: 10.13005/ojc/340128
3. Yu. N. Sazanov, *Russ. J. Appl. Chem.*, **2001**, *74*, 1253–1269. DOI: 10.1023/A:1013768725369
4. G. Sun, L. Liu, J. Wang, H. Wang, Z. Xie, S. Han, *Polym. Degrad. Stab.*, **2014**, *110*, 1–12. DOI: 10.1016/j.polymdegradstab.2014.08.011
5. X.-H. Ren, G.-H. Sun, L.-C. Wang, R.-R. Chen, J. Wang, S.-H. Han, *Chin. J. Polym. Sci.*, **2021**, *39*, 237–248. DOI: 10.1007/s10118-020-2482-y
6. E. Gubbelmans, T. Verbiest, M. Van Beylen, A. Persoons, C. Samyn, *Polymer*, **2002**, *43*, 1581–1585. DOI: 10.1016/S0032-3861(01)00678-4
7. K. K. K. Swamy, N. N. B. Kumar, E. Balaraman, K. V. P. P. Kumar, *Chem. Rev.*, **2009**, *109*, 2551–2651. DOI: 10.1021/cr800278z
8. G.-S. Liou, S.-H. Hsiao, M. Ishida, M. Kakimoto, Y. Imai, *J. Polym. Sci., Part A: Polym. Chem.*, **2002**, *40*, 3815–3822. DOI: 10.1002/pola.10430
9. C. Gao, X. Wu, G. Lv, M. Ding, L. Gao, *Macromolecules*, **2004**, *37*, 2754–2761. DOI: 10.1021/ma0350720
10. R. J. Perry, S. E. Tunney, B. D. Wilson, *Macromolecules*, **1996**, *29*, 1014–1020. DOI: 10.1021/ma950890g
11. L. Schmitz, M. Rehahn, M. Ballauff, *Polymer*, **1993**, *34*, 646–649. DOI: 10.1016/0032-3861(93)90565-R
12. R. O. Johnson, H. S. Burlhis, *J. Polym. Sci., Polym. Symp.*, **1983**, *70*, 129–143. DOI: 10.1002/polc.5070700111
13. H.-C. Yu, S. V. Kumar, Y.-K. Song, J. Choi, K. Kudo, J.-G. Kim, S.-Y. Oh, C.-M. Chung, *Macromol. Res.*, **2011**, *19*, 1272–1277. DOI: 10.1007/s13233-011-1216-y
14. A. S. Shaplov, D. O. Ponkratov, Y. S. Vygodskii, *Polym. Sci., Ser. B*, **2016**, *58*, 73–142. DOI: 10.1134/S156009041602007X
15. S. Mallakpour, M. Dinari, *Iran. Polym. J.*, **2011**, *20*, 259–279.
16. H. Frank, U. Ziener, K. Landfester, *Macromolecules*, **2009**, *42*, 7846–7853. DOI: 10.1021/ma901392h
17. E. E. Said-Galiyev, Y. S. Vygodskii, L. N. Nikitin, R. A. Vinokur, M. O. Gallyamov, I. V. Pototskaya, V. V. Kireev, A. R. Khokhlov, K. Schaumburg, *J. Supercrit. Fluids*, **2003**, *26*, 147–156. DOI: 10.1016/S0896-8446(02)00210-3



18. E. Said-Galiyev, I. Pototskaya, Y. S. Vygodskii, *Polym. Sci., Ser. C: Rev.*, **2004**, *46*, 1–13.
19. E. E. Said-Galiev, M. L. Keshtov, A. R. Khokhlov, Ya. S. Vygodskii, R. A. Dvorikova, N. M. Belomoina, E. G. Bulychева, *Russ. Chem. Bull.*, **2020**, *69*, 1035–1053. DOI: 10.1007/s11172-020-2865-9
20. M. Haruki, N. Fukui, S.-i. Kihara, S. Takishima, *J. Chem. Thermodyn.*, **2012**, *54*, 261–265. DOI: 10.1016/j.jct.2012.04.013
21. RU Patent 2270842, **2006**.
22. E. S. Alekseev, A. Yu. Alentiev, A. S. Belova, V. I. Bogdan, T. V. Bogdan, A. V. Bystrova, E. R. Gafarova, E. N. Golubeva, E. A. Grebenik, O. I. Gromov, V. A. Davankov, S. G. Zlotin, M. G. Kiselev, A. E. Koklin, Y. N. Kononevich, A. E. Lazhko, V. V. Lunin, S. E. Lyubimov, O. N. Martyanov, I. I. Mishanin, A. M. Muzafarov, N. S. Nesterov, A. Yu. Nikolaev, R. D. Oparin, O. O. Parenago, O. P. Parenago, Y. A. Pokusaeva, I. A. Ronova, A. B. Solovieva, M. N. Temnikov, P. S. Timashev, O. V. Turova, E. V. Filatova, A. A. Philippov, A. M. Chibiryaev, A. S. Shalygin, *Russ. Chem. Rev.*, **2020**, *89*, 1337–1427. DOI: 10.1070/rcr4932
23. Y. S. Vygodskii, E. I. Lozinskaya, A. S. Shaplov, *Macromol. Rapid Commun.*, **2002**, *23*, 676–680. DOI: 10.1002/1521-3927(20020801)23:12<676::AID-MARC676>3.0.CO;2-2
24. Y. S. Vygodskii, E. I. Lozinskaya, A. S. Shaplov, K. A. Lyssenko, M. Yu. Antipin, Y. G. Urman, *Polymer*, **2004**, *45*, 5031–5045. DOI: 10.1016/j.polymer.2004.05.025
25. S. K. Singh, A. W. Savoy, *J. Mol. Liq.*, **2020**, *297*, 112038. DOI: 10.1016/j.molliq.2019.112038
26. E. Gharekhani, A. S. Mehdipour, G. M. Taghi, *Iran. Polym. J.*, **2011**, *20*, 491–502.
27. J. Szala-Bilnik, A. Abedini, E. Crabtree, J. E. Bara, C. H. Turner, *J. Phys. Chem. B*, **2019**, *123*, 7455–7463. DOI: 10.1021/acs.jpcc.9b05555
28. R. D. Rusu, M. J. Abadie, in: *Polyimide for Electronic and Electrical Engineering Applications*, S. Diahm (Ed.), IntechOpen, London, **2020**, ch. 15. DOI: 10.5772/intechopen.93340
29. A. S. Shaplov, S. M. Morozova, E. I. Lozinskaya, P. S. Vlasov, A. S. Gouveia, L. C. Tomé, I. M. Marrucho, Y. S. Vygodskii, *Polym. Chem.*, **2016**, *7*, 580–591. DOI: 10.1039/c5py01553g
30. A. A. Kuznetsov, *High Perform. Polym.*, **2000**, *12*, 445–460. DOI: 10.1088/0954-0083/12/3/307
31. A. A. Kuznetsov, A. Yu. Tsegelskaya, A. M. Orlova, N. A. Belov, S. V. Chirkov, R. Yu. Nikiforov, A. Yu. Alentiev, *Membr. Technol.*, **2019**, *1*, 316–322. DOI: 10.1134/S2517751619050044
32. A. A. Kuznetsov, M. R. Batuashvili, V. Kaminskii, *High Perform. Polym.*, **2017**, *29*, 716–723. DOI: 10.1177/0954008317702953
33. F. Hasanain, Z. Y. Wang, *Polymer*, **2008**, *49*, 831–835. DOI: 10.1016/j.polymer.2007.11.058
34. M. M. Unterlass, D. Kopetzki, M. Antonietti, J. Weber, *Polym. Chem.*, **2011**, *2*, 1744–1753. DOI: 10.1039/c1py00109d
35. B. Baumgartner, M. J. Bojdys, P. Skrinjar, M. M. Unterlass, *Macromol. Chem. Phys.*, **2016**, *217*, 485–500. DOI: 10.1002/macp.201500287
36. M. J. Taublaender, M. Reiter, M. M. Unterlass, *Macromolecules*, **2019**, *52*, 6318–6329. DOI: 10.1021/acs.macromol.9b00985
37. M. E. Barley, A. L. Pickard, S. G. Hagemann, S. L. Folkert, *Miner. Deposita*, **1999**, *34*, 784–789. DOI: 10.1007/s001260050238
38. B. Baumgartner, M. Puchberger, M. M. Unterlass, *Polym. Chem.*, **2015**, *6*, 5773–5781. DOI: 10.1039/c5py00231a
39. B. Baumgartner, M. J. Bojdys, M. M. Unterlass, *Polym. Chem.*, **2014**, *5*, 3771–3776. DOI: 10.1039/c4py00263f
40. Z. Cui, J. Zhou, T. Liu, Y. Wang, Y. Hu, Y. Wang, Z. Zou, *Chem. Asian J.*, **2019**, *14*, 2138–2148. DOI: 10.1002/asia.201900261
41. S. Chu, Y. Hu, J. Zhang, Z. Cui, J. Shi, Y. Wang, Z. Zou, *Int. J. Hydrogen Energy*, **2021**, *46*, 9064–9076. DOI: 10.1016/j.ijhydene.2020.12.225
42. M. M. Unterlass, F. Emmerling, M. Antonietti, J. Weber, *Chem. Commun.*, **2014**, *50*, 430–432. DOI: 10.1039/c3cc47674j
43. L. Zhou, Y. Li, Z. Wang, M. Zhang, X. Wang, H. Niu, D. Wu, *RSC Adv.*, **2019**, *9*, 7314–7320. DOI: 10.1039/c9ra00355j
44. K. Cheng, J.-p. Hu, Y.-c. Wu, C.-q. Shi, Z.-g. Chen, S.-m. Liu, Y.-c. Yuan, Y.-q. Mo, J.-q. Zhao, *Royal Soc. Open Sci.*, **2019**, *6*, 190196. DOI: 10.1098/rsos.190196
45. J.-Y. Choi, S.-W. Jin, D.-M. Kim, I.-H. Song, K.-N. Nam, H.-J. Park, C.-M. Chung, *Polymers*, **2019**, *11*, 477. DOI: 10.3390/polym11030477
46. S. M. Budy, J. D. Hall, D. Y. Son, *Polym. Chem.*, **2020**, *11*, 6273–6280. DOI: 10.1039/d0py00814a
47. L. Zhuo, S. Tang, K. Zhao, F. Xie, Y. Bai, *J. Appl. Polym. Sci.*, **2020**, *137*, 48484. DOI: 10.1002/app.48484
48. B. Schartel, J. H. Wendorff, *Polym. Eng. Sci.*, **1999**, *39*, 128–151. DOI: 10.1002/pen.11403
49. V. Neverov, S. Chvalun, J. Blackwell, F. Harris, S. D. Cheng, *Polym. Sci., Ser. A*, **2000**, *42*, 299–308.
50. G. Pawlikowski, D. Dutta, R. A. Weiss, *Annu. Rev. Mater. Sci.*, **1991**, *21*, 159–184. DOI: 10.1146/annurev.ms.21.080191.001111
51. O. N. Zabegaeva, D. A. Sapozhnikov, Ya. S. Vygodskii, *Polym. Sci., Ser. C*, **2020**, *62*, 183–195. DOI: 10.1134/s1811238220020174
52. M. T. DeMeuse, G. Kiss, in: *High Temperature Polymer Blends*, M. T. DeMeuse (Ed.), Elsevier, Cambridge, **2014**, ch. 5, pp. 141–164. DOI: 10.1533/9780857099013.141
53. W. F. Yong, H. Zhang, *Prog. Mater. Sci.*, **2021**, *116*, 100713. DOI: 10.1016/j.pmatsci.2020.100713
54. *Handbook of Polymer Blends and Composites*, A. K. Kulshreshtha, C. Vasile (Eds.), iSmithers Rapra Publ., Shrewsbury, **2002**.
55. *Polymer Blends and Alloys*, M. Folkes, P. S. Hope (Eds.), Springer, London, **1993**.
56. H. A. Mannan, H. Mukhtar, T. Murugesan, R. Nasir, D. F. Mohshim, A. Mushtaq, *Chem. Eng. Technol.*, **2013**, *36*, 1838–1846. DOI: 10.1002/ceat.201300342
57. C. A. Scholes, C. P. Ribeiro, S. E. Kentish, B. D. Freeman, *Sep. Purif. Technol.*, **2014**, *124*, 134–140. DOI: 10.1016/j.seppur.2014.01.012
58. M. G. García, J. Marchese, N. A. Ochoa, *Int. J. Hydrogen Energy*, **2010**, *35*, 8983–8992. DOI: 10.1016/j.ijhydene.2010.06.038
59. H. Ke, L. Zhao, X. Zhang, Y. Qiao, G. Wang, X. Wang, *Polym. Test.*, **2020**, *90*, 106746. DOI: 10.1016/j.polymertesting.2020.106746
60. S. Niyogi, S. Maiti, B. Adhikari, *Polym. Degrad. Stab.*, **2000**, *68*, 459–464. DOI: 10.1016/S0141-3910(00)00037-9
61. S. Niyogi, S. Maiti, B. Adhikari, *Eur. Polym. J.*, **2001**, *37*, 2079–2089. DOI: 10.1016/S0014-3057(01)00098-2
62. S. Niyogi, S. Maiti, B. Adhikari, *Polym. Eng. Sci.*, **2002**, *42*, 336–345. DOI: 10.1002/pen.10952
63. I. Butnaru, C.-P. Constantin, M. Asandulesa, A. Wolińska-Grabczyk, A. Jankowski, U. Szeluga, M.-D. Damaceanu, *Sep. Purif. Technol.*, **2020**, *233*, 116031. DOI: 10.1016/j.seppur.2019.116031
64. A. Kushwaha, M. E. Dose, S. Luo, B. D. Freeman, R. Guo, *Sep. Purif. Technol.*, **2017**, *184*, 384–393. DOI: 10.1016/j.seppur.2017.04.051
65. M. A. Abd. Hamid, Y. T. Chung, R. Rohani, Mohd. U. Mohd. Junaidi, *Sep. Purif. Technol.*, **2019**, *209*, 598–607. DOI: 10.1016/j.seppur.2018.07.067
66. S. Rafiq, Z. Man, S. Maitra, A. Maulud, F. Ahmad, N. Muhammad, *Korean J. Chem. Eng.*, **2011**, *28*, 2050–2056. DOI: 10.1007/s11814-011-0053-1
67. G. C. Kapantaidakis, G. H. Koops, *J. Membr. Sci.*, **2002**, *204*, 153–171. DOI: 10.1016/S0376-7388(02)00030-3
68. L. Escorial, M. de la Viuda, S. Rodríguez, A. Tena, A. Marcos, L. Palacio, P. Prádanos, A. E. Lozano, A. Hernández, *Eur. Polym. J.*, **2018**, *103*, 390–399. DOI: 10.1016/j.eurpolymj.2018.04.031

69. M. Zhang, G. Wang, A. Li, X. Wei, F. Li, J. Zhang, J. Chen, R. Wang, *J. Membr. Sci.*, **2021**, *619*, 118800. DOI: 10.1016/j.memsci.2020.118800
70. H. Sanaeepur, A. E. Amooghin, S. Bandehali, A. Moghadassi, T. Matsuura, B. Van der Bruggen, *Prog. Polym. Sci.*, **2019**, *91*, 80–125. DOI: 10.1016/j.progpolymsci.2019.02.001
71. D. Ljubic, W. Liu, N.-X. Hu, Y. Wu, S. Zhu, *Org. Electronics*, **2018**, *59*, 349–357. DOI: 10.1016/j.orgel.2018.05.036
72. S. Mazinani, S. Darvishmanesh, R. Ramazani, B. Van der Bruggen, *React. Funct. Polym.*, **2017**, *111*, 88–101. DOI: 10.1016/j.reactfunctpolym.2016.12.010
73. T. Takeichi, Y. Guo, S. Rimdusit, *Polymer*, **2005**, *46*, 4909–4916. DOI: 10.1016/j.polymer.2005.03.096
74. S. Rimdusit, S. Tiptipakorn, C. Jubsilp, T. Takeichi, *React. Funct. Polym.*, **2013**, *73*, 369–380. DOI: 10.1016/j.reactfunctpolym.2012.04.022
75. H. Ishida, in: *Handbook of Benzoxazine Resins*, H. Ishida, T. Agag (Eds.), Elsevier, Oxford, **2011**, part I, ch. 1, pp. 3–81. DOI: 10.1016/B978-0-444-53790-4.00046-1
76. T. Takeichi, T. Kawachi, T. Agag, in: *Handbook of Benzoxazine Resins*, H. Ishida, T. Agag (Eds.), Elsevier, Oxford, **2011**, part VI, ch. 20, pp. 378–387. DOI: 10.1016/B978-0-444-53790-4.00065-5
77. T. Takeichi, T. Agag, *High Perform. Polym.*, **2016**, *18*, 777–797. DOI: 10.1177/0954008306068254
78. K. Zhang, J. Liu, H. Ishida, *RSC Adv.*, **2014**, *4*, 62550–62556. DOI: 10.1039/c4ra12015a
79. T. Takeichi, T. Agag, R. Zeidam, *J. Polym. Sci., Part A: Polym. Chem.*, **2001**, *39*, 2633–2641. DOI: 10.1002/pola.1240
80. S. Tiptipakorn, S. Damrongsakkul, S. Ando, K. Hemvichian, S. Rimdusit, *Polym. Degrad. Stab.*, **2007**, *92*, 1265–1278. DOI: 10.1016/j.polymdegradstab.2007.03.021
81. S. Miyata, K. Yoshida, H. Shirokura, M. Kashio, K. Nagai, *Polym. Int.*, **2009**, *58*, 1148–1159. DOI: 10.1002/pi.2643
82. S. Sato, M. Ichikawa, T. Ose, S. Miyata, Y. Takahashi, S. Kanehashi, H. Matsumoto, K. Nagai, *Polym. Int.*, **2013**, *62*, 1377–1385. DOI: 10.1002/pi.4433
83. S. Kanehashi, Y. Koyama, S. Ando, S. Konishi, R. Shindo, S. Miyata, S. Sato, T. Miyakoshi, K. Nagai, *Polym. Int.*, **2014**, *63*, 435–444. DOI: 10.1002/pi.4520
84. T. Suzuki, A. Yoshida, S. Ando, K. Nagai, *Polym. Int.*, **2015**, *64*, 1209–1218. DOI: 10.1002/pi.4897
85. A. Yoshida, K. Nagai, 252nd ACS Nat. Meet. Expo., Philadelphia, USA, **2016**; PMSE-490, Philadelphia, USA, **2016**.
86. N. Taniguchi, 252nd ACS Nat. Meet. Expo., Philadelphia, USA, **2016**.
87. S. Ando, Y. Koyama, S. Miyata, S. Sato, S. Kanehashi, K. Nagai, *Polym. Int.*, **2014**, *63*, 1634–1642. DOI: 10.1002/pi.4681
88. S. Ando, A. Yoshida, K. Nagai, *Polym. Eng. Sci.*, **2016**, *56*, 1191–1200. DOI: 10.1002/pen.24353
89. Y. Sasago, 252nd ACS Nat. Meet. Expo., Philadelphia, USA, **2016**; PMSE-449, Philadelphia, USA, **2016**.
90. S. Ando, A. Yoshida, K. Nagai, *J. Appl. Polym. Sci.*, **2015**, *132*, 42208. DOI: 10.1002/app.42208
91. F. J. Carrière, H. Sekiguchi, N. N. Surin, V. A. Kotelnikov, Ya. S. Vygodskii, *Polym. Bull.*, **1995**, *35*, 441–448. DOI: 10.1007/BF00297610
92. Y. Pae, F. W. Harris, *J. Polym. Sci., Part A: Polym. Chem.*, **2000**, *38*, 4247–4257. DOI: 10.1002/1099-0518(20001201)38:23<4247::AID-POLA100>3.0.CO;2-8
93. Y. Pae, *J. Appl. Polym. Sci.*, **2006**, *99*, 300–308. DOI: 10.1002/app.22480
94. Y. Pae, *J. Appl. Polym. Sci.*, **2006**, *99*, 309–318. DOI: 10.1002/app.22481
95. G. D. Fu, E. T. Kang, K. G. Neoh, C. C. Lin, D. J. Liaw, *Macromolecules*, **2005**, *38*, 7593–7600. DOI: 10.1021/ma0506435
96. A. V. Yakimanskii, T. K. Meleshko, D. M. Il'gach, N. N. Bogorad, E. N. Vlasova, T. D. Anan'eva, *Russ. Chem. Bull.*, **2012**, *61*, 999–1008. DOI: 10.1007/s11172-012-0129-z
97. A. P. Filippov, E. V. Belyaeva, T. K. Meleshko, A. V. Yakimanskii, *J. Polym. Sci., Part B: Polym. Phys.*, **2014**, *52*, 1539–1546. DOI: 10.1002/polb.23595
98. T. K. Meleshko, A. S. Ivanova, A. V. Kashina, I. V. Ivanov, T. N. Nekrasova, N. V. Zakharova, A. P. Filippov, A. V. Yakimanskii, *Polym. Sci., Ser. B*, **2017**, *59*, 674–688. DOI: 10.1134/s1560090417060045
99. T. K. Meleshko, A. Yu. Pulyalina, N. S. Tyan, G. A. Polotskaya, I. V. Ivanov, N. V. Kukarkina, A. M. Toikka, A. V. Yakimanskii, *Polym. Sci., Ser. B*, **2017**, *59*, 183–193. DOI: 10.1134/s1560090417020063
100. T. K. Meleshko, I. V. Ivanov, A. V. Kashina, N. N. Bogorad, M. A. Simonova, N. V. Zakharova, A. P. Filippov, A. V. Yakimanskii, *Polym. Sci., Ser. B*, **2018**, *60*, 35–50. DOI: 10.1134/s1560090418010098
101. V. D. Pautov, T. N. Nekrasova, T. D. Anan'eva, T. K. Meleshko, D. M. Ilgach, A. V. Yakimanskii, *Polym. Sci., Ser. A*, **2013**, *55*, 526–534. DOI: 10.1134/s0965545x13080051
102. A. V. Yakimanskii, T. K. Meleshko, D. M. Ilgach, M. A. Bauman, T. D. Anan'eva, L. G. Klapshina, S. A. Lermontova, I. V. Balalaeva, W. E. Douglas, *J. Polym. Sci., Part A: Polym. Chem.*, **2013**, *51*, 4267–4281. DOI: 10.1002/pola.26846
103. A. P. Filippov, E. V. Belyaeva, N. V. Zakharova, A. S. Sasina, D. M. Ilgach, T. K. Meleshko, A. V. Yakimanskii, *Colloid Polym. Sci.*, **2015**, *293*, 555–565. DOI: 10.1007/s00396-014-3441-3
104. M. Higa, K. Yaguchi, R. Kitani, *Electrochim. Acta*, **2010**, *55*, 1380–1384. DOI: 10.1016/j.electacta.2009.07.046
105. A. V. Kashina, T. K. Meleshko, N. N. Bogorad, M. A. Bezrukova, A. V. Yakimanskii, *Polym. Sci., Ser. C*, **2019**, *61*, 174–185. DOI: 10.1134/s1811238219010090
106. V. D. Pautov, T. N. Nekrasova, T. D. Anan'eva, T. K. Meleshko, I. V. Ivanov, A. V. Yakimanskii, *J. Polym. Res.*, **2018**, *25*, 8. DOI: 10.1007/s10965-017-1404-3
107. S. A. Klimova, O. A. Inozemtseva, S. V. German, D. A. Gorin, D. M. Ilgach, T. K. Meleshko, A. V. Yakimanskii, *Prot. Met. Phys. Chem. Surf.*, **2015**, *51*, 396–406. DOI: 10.1134/s2070205115030132
108. Y. S. Vygodskii, T. V. Volkova, T. L. Batalova, O. N. Zabegaeva, E. M. Belavtseva, A. A. Sakharova, R. G. Gasanov, D. A. Sapozhnikov, V. Yu. Voytekunas, *High Perform. Polym.*, **2009**, *21*, 579–595. DOI: 10.1177/0954008309339410
109. T. V. Volkova, Y. S. Vygodskii, O. N. Zabegaeva, Y. V. Zubavichus, M. N. Il'ina, A. P. Krasnov, O. V. Afonicheva, E. I. Lozinskaya, I. A. Garbuzova, A. S. Shaplov, *J. Appl. Polym. Sci.*, **2009**, *114*, 577–586. DOI: 10.1002/app.30543
110. O. N. Zabegaeva, G. Chen, X. Fang, T. M. Aliev, Ya. S. Vygodskii, *Polym. Sci., Ser. B*, **2020**, *62*, 94–101. DOI: 10.1134/s1560090420020098
111. Y. S. Vygodskii, A. M. Matieva, A. A. Sakharova, D. A. Sapozhnikov, T. V. Volkova, *High Perform. Polym.*, **2001**, *13*, S317–S326. DOI: 10.1088/0954-0083/13/2/327
112. M. J. M. Abadie, V. Y. Voytekunas, A. Matieva, Y. S. Vygodskii, *Polym. Sci., Ser. B*, **2003**, *45*, 67–70.
113. Y. S. Vygodskii, T. V. Volkova, A. A. Sakharova, D. A. Sapozhnikov, A. Matieva, *Polym. Sci., Ser. A*, **2002**, *44*, 1249–1254.
114. D. A. Sapozhnikov, T. V. Volkova, A. A. Sakharova, R. G. Gasanov, V. Yu. Voytekunas, M. J. M. Abadie, J.-Y. Sanchez, Y. S. Vygodskii, *Int. J. Polym. Sci.*, **2009**, *2009*, 527046. DOI: 10.1155/2009/527046
115. D. A. Sapozhnikov, T. V. Volkova, A. A. Sakharova, R. G. Gasanov, V. Yu. Voytekunas, M. J. M. Abadie, J.-Y. Sanchez, Ya. S. Vygodskii, *Polym. Sci., Ser. B*, **2009**, *51*, 1–12. DOI: 10.1134/s1560090409010011
116. Y. S. Vygodskii, A. A. Sakharova, A. M. Matieva, *High Perform. Polym.*, **1999**, *11*, 379–386. DOI: 10.1088/0954-0083/11/4/303
117. Y. S. Vygodskii, A. Sakharova, A. Matieva, *Polym. Sci., Ser. B*, **2000**, *42*, 62–64.

118. Ya. S. Vygodskii, T. V. Volkova, A. A. Sakharova, D. A. Sapozhnikov, G. G. Nikiforova, A. M. Matieva, *Polym. Sci., Ser. A*, **2006**, *48*, 683–688. DOI: 10.1134/s0965545x06070030
119. Y. S. Vygodskii, A. Matieva, T. Volkova, A. Sakharova, D. Sapozhnikov, *Polym. Sci., Ser. A*, **2004**, *46*, 352–360.
120. D. A. Sapozhnikov, B. A. Baiminov, Ya. S. Vygodskii, *Dokl. Chem.*, **2016**, *468*, 169–173. DOI: 10.1134/s0012500816040066
121. Y. S. Vygodskii, D. A. Sapozhnikov, B. A. Bayminov, S. L. Semjonov, A. F. Kosolapov, E. A. Plastinin, *Prog. Org. Coat.*, **2016**, *99*, 210–215. DOI: 10.1016/j.porgcoat.2016.05.023
122. S. Niyogi, M. Hamon, H. Hu, B. Zhao, P. Bhowmik, R. Sen, M. Itkis, R. Haddon, *Acc. Chem. Res.*, **2002**, *35*, 1105–1113. DOI: 10.1021/ar010155r
123. A. V. Krestinin, A. P. Kharitonov, *Polym. Sci., Ser. B*, **2018**, *60*, 516–529. DOI: 10.1134/S156009041804005X
124. M. Hegde, U. Lafont, B. Norder, S. J. Picken, E. T. Samulski, M. Rubinstein, T. Dingemans, *Macromolecules*, **2013**, *46*, 1492–1503. DOI: 10.1021/ma302456h
125. W. Huang, Y. Lin, S. Taylor, J. Gaillard, A. M. Rao, Y.-P. Sun, *Nano Lett.*, **2002**, *2*, 231–234. DOI: 10.1021/nl010083x
126. L. Qu, Y. Lin, D. E. Hill, B. Zhou, W. Wang, X. Sun, A. Kitaygorodskiy, M. Suarez, J. W. Connell, L. F. Allard, Y.-P. Sun, *Macromolecules*, **2004**, *37*, 6055–6060. DOI: 10.1021/ma0491006
127. J. J. Ge, D. Zhang, Q. Li, H. Hou, M. J. Graham, L. Dai, F. W. Harris, S. Z. D. Cheng, *J. Am. Chem. Soc.*, **2005**, *127*, 9984–9985. DOI: 10.1021/ja050924s
128. D. Hill, Y. Lin, L. Qu, A. Kitaygorodskiy, J. W. Connell, L. F. Allard, Y.-P. Sun, *Macromolecules*, **2005**, *38*, 7670–7675. DOI: 10.1021/ma0509210
129. K. E. Wise, C. Park, E. J. Siochi, J. S. Harrison, *Chem. Phys. Lett.*, **2004**, *391*, 207–211. DOI: 10.1016/j.cplett.2004.04.096
130. Z. Ounaies, C. Park, K. E. Wise, E. J. Siochi, J. S. Harrison, *Compos. Sci. Technol.*, **2003**, *63*, 1637–1646. DOI: 10.1016/s0266-3538(03)00067-8
131. S.-M. Yuen, C.-C. M. Ma, C.-L. Chiang, Y.-Y. Lin, C.-C. Teng, *J. Polym. Sci., Part A: Polym. Chem.*, **2007**, *45*, 3349–3358. DOI: 10.1002/pola.22085
132. T. Liu, Y. Tong, W.-D. Zhang, *Compos. Sci. Technol.*, **2007**, *67*, 406–412. DOI: 10.1016/j.compscitech.2006.09.007
133. B. Zhang, M.-H. Lee, A. N. Chakoli, W. Zang, K. Zhang, Y. Zhang, G. Song, C. Chen, X. Li, Y. Li, *RSC Adv.*, **2014**, *4*, 14024–14030. DOI: 10.1039/c3ra43977a
134. B. Zhang, A. N. Chakoli, W. Zang, Y. Tian, K. Zhang, C. Chen, Y. Li, *J. Appl. Polym. Sci.*, **2014**, *131*, 40479. DOI: 10.1002/app.40479
135. B. Zhou, Y. Lin, D. E. Hill, W. Wang, L. M. Veca, L. Qu, P. Pathak, M. J. Meziani, J. Diaz, J. W. Connell, K. A. Watson, L. F. Allard, Y.-P. Sun, *Polymer*, **2006**, *47*, 5323–5329. DOI: 10.1016/j.polymer.2006.05.055
136. W. Ning, Z. Wang, P. Liu, D. Zhou, S. Yang, J. Wang, Q. Li, S. Fan, K. Jiang, *Carbon*, **2018**, *139*, 1136–1143. DOI: 10.1016/j.carbon.2018.08.011
137. V. Georgakilas, M. Otyepka, A. B. Bourlinos, V. Chandra, N. Kim, K. C. Kemp, P. Hobza, R. Zboril, K. S. Kim, *Chem. Rev.*, **2012**, *112*, 6156–6214. DOI: 10.1021/cr3000412
138. L. Ma, G. Wang, J. Dai, *High Perform. Polym.*, **2016**, *29*, 187–196. DOI: 10.1177/09540083166634177
139. J. Zhu, C.-H. Lee, H.-I. Joh, H. C. Kim, S. Lee, *Carbon Lett.*, **2012**, *13*, 230–235. DOI: 10.5714/cl.2012.13.4.230
140. K.-C. Ke, C. Cheng, L.-J. Lin, S.-Y. Yang, *Microsyst. Technol.*, **2018**, *24*, 3283–3289. DOI: 10.1007/s00542-018-3824-3
141. Z. Song, T. Xu, M. L. Gordin, Y.-B. Jiang, I.-T. Bae, Q. Xiao, H. Zhan, J. Liu, D. Wang, *Nano Lett.*, **2012**, *12*, 2205–2211. DOI: 10.1021/nl2039666
142. Y. Huang, K. Li, J. Liu, X. Zhong, X. Duan, I. Shakir, Y. Xu, *J. Mater. Chem. A*, **2017**, *5*, 2710–2716. DOI: 10.1039/c6ta09754e
143. T. Huang, Y. Xin, T. Li, S. Nutt, C. Su, H. Chen, P. Liu, Z. Lai, *ACS Appl. Mater. Interfaces*, **2013**, *5*, 4878–4891. DOI: 10.1021/am400635x
144. X. Wu, Z. Jin, Y. Zhu, H. Zhao, *High Perform. Polym.*, **2020**, *33*, 40–51. DOI: 10.1177/0954008320940358
145. X. Wu, Y. Zhang, P. Du, Z. Jin, H. Zhao, L. Wang, *New J. Chem.*, **2019**, *43*, 5697–5705. DOI: 10.1039/c9nj00216b
146. K. C. Chang, C. H. Hsu, H. I. Lu, W. F. Ji, C. H. Chang, W. Y. Li, T. L. Chuang, J. M. Yeh, W. R. Liu, M. H. Tsai, *EXPRESS Polym. Lett.*, **2014**, *8*, 243–255. DOI: 10.3144/expresspolymlett.2014.28
147. M.-H. Tsai, I.-H. Tseng, Y.-C. Huang, H.-P. Yu, P.-Y. Chang, *Adv. Eng. Mater.*, **2016**, *18*, 582–590. DOI: 10.1002/adem.201500286
148. P. Zhang, J. Zhao, K. Zhang, R. Bai, Y. Wang, C. Hua, Y. Wu, X. Liu, H. Xu, Y. Li, *Compos. Part A: Appl. Sci. Manuf.*, **2016**, *84*, 428–434. DOI: 10.1016/j.compositesa.2016.02.019
149. X. Chen, H. Huang, X. Shu, S. Liu, J. Zhao, *RSC Adv.*, **2017**, *7*, 1956–1965. DOI: 10.1039/c6ra25343a
150. P. Ma, C. Dai, H. Wang, Z. Li, H. Liu, W. Li, C. Yang, *Compos. Commun.*, **2019**, *16*, 84–93. DOI: 10.1016/j.coco.2019.08.011
151. J. Yang, Y. Ye, X. Li, X. Lü, R. Chen, *Compos. Sci. Technol.*, **2018**, *164*, 187–194. DOI: 10.1016/j.compscitech.2018.05.044
152. B. Feng, K. Xu, A. Huang, *RSC Adv.*, **2017**, *7*, 2211–2217. DOI: 10.1039/c6ra24974d
153. A. Kausar, *Polym.-Plast. Technol. Eng.*, **2017**, *56*, 1375–1390. DOI: 10.1080/03602559.2016.1275688
154. G. Kickelbick, *Prog. Polym. Sci.*, **2003**, *28*, 83–114. DOI: 10.1016/S0079-6700(02)00019-9
155. H. II Shin, J.-H. Chang, *Polymers*, **2020**, *12*, 135. DOI: 10.3390/polym12010135
156. Z.-M. Liang, J. Yin, J.-H. Wu, Z.-X. Qiu, F.-F. He, *Eur. Polym. J.*, **2004**, *40*, 307–314. DOI: 10.1016/j.eurpolymj.2003.09.020
157. T.-Y. Chao, H.-L. Chang, W.-C. Su, J.-Y. Wu, R.-J. Jeng, *Dyes Pigm.*, **2008**, *77*, 515–524. DOI: 10.1016/j.dyepig.2007.08.001
158. M. Tommalieh, A. Zihlif, *Physica B: Condens. Mater.*, **2010**, *405*, 4750–4754. DOI: 10.1016/j.physb.2010.08.070
159. C. Huang, J. Li, G. Xie, F. Han, D. Huang, F. Zhang, B. Zhang, G. Zhang, R. Sun, C.-P. Wong, *Macromol. Mater. Eng.*, **2019**, *304*, 1900505. DOI: 10.1002/mame.201900505
160. X. Liu, J. Yin, Y. Kong, M. Chen, Y. Feng, K. Yan, X. Li, B. Su, Q. Lei, *Thin Solid Films*, **2013**, *544*, 352–356. DOI: 10.1016/j.tsf.2013.02.100
161. N. Atar, E. Grossman, I. Gouzman, A. Bolker, V. J. Murray, B. C. Marshall, M. Qian, T. K. Minton, Y. Hanein, *ACS Appl. Mater. Interfaces*, **2015**, *7*, 12047–12056. DOI: 10.1021/acsami.5b02200
162. G. Song, X. Li, Q. Jiang, J. Mu, Z. Jiang, *RSC Adv.*, **2015**, *5*, 11980–11988. DOI: 10.1039/c4ra14727h
163. Y. W. Liu, J. J. Liu, Q. Ding, J. H. Tan, J. Y. Chen, Z. H. She, A. Tang, K. J. Zeng, *Polym. Compos.*, **2018**, *39*, E1725–E1732. DOI: 10.1002/pc.24713
164. S. M. Davoodi, M. Sadeghi, M. Naghsh, A. Moheb, *RSC Adv.*, **2016**, *6*, 23746–23759. DOI: 10.1039/c6ra00553e
165. S. Rafiq, Z. Man, A. Maulud, N. Muhammad, S. Maitra, *Sep. Purif. Technol.*, **2012**, *90*, 162–172. DOI: 10.1016/j.seppur.2012.02.031
166. A. C. Lua, Y. Shen, *Chem. Eng. J.*, **2013**, *220*, 441–451. DOI: 10.1016/j.cej.2012.11.140
167. W. J. Bae, M. K. Kovalev, F. Kalinina, M. Kim, C. Cho, *Polymer*, **2016**, *105*, 124–132. DOI: 10.1016/j.polymer.2016.10.023
168. A. S. Hicyilmaz, A. C. Bedeloglu, *SN Appl. Sci.*, **2021**, *3*, 363. DOI: 10.1007/s42452-021-04362-5
169. H. Ahmadizadegan, S. Esmailzadeh, *Int. J. Polym. Anal. Charact.*, **2020**, *25*, 604–620. DOI: 10.1080/1023666X.2020.1827529
170. H. H. Yong, H. C. Park, Y. S. Kang, J. Won, W. N. Kim, *J. Membr. Sci.*, **2001**, *188*, 151–163. DOI: 10.1016/S0376-7388(00)00659-1
171. S. Sinha Ray, M. Okamoto, *Prog. Polym. Sci.*, **2003**, *28*, 1539–1641. DOI: 10.1016/j.progpolymsci.2003.08.002

172. V. Mittal, *Materials*, **2009**, *2*, 992–1057. DOI: 10.3390/ma2030992
173. T. Tereshchenko, *Polym. Sci., Ser. B*, **2008**, *50*, 249–262. DOI: 10.1134/S1560090408090054
174. H. Birtane, K. Esmer, S. Madakbas, M. V. Kahraman, *J. Macromol. Sci., Part A*, **2019**, *56*, 245–252. DOI: 10.1080/10601325.2019.1565546
175. M. G. Mohamed, S. W. Kuo, *Polymers*, **2019**, *11*, 26. DOI: 10.3390/polym11010026
176. N. I. Cherkashina, P. V. Matyuhin, R. N. Yastrebinsky, Z. V. Pavlenko, O. V. Demchenko, *Int. J. Appl. Fundam. Res.*, **2015**, 991–994.
177. N. I. Cherkashina, V. I. Pavlenko, O. V. Demchenko, D. A. Ivanitsky, *Adv. Curr. Nat. Sci.*, **2015**, 524–527.
178. A. D. Breki, A. L. Didenko, V. V. Kudryavtsev, E. S. Vasilyeva, O. V. Tolochko, A. G. Kolmakov, A. E. Gvozdev, D. A. Provotorov, N. E. Starikov, Yu. A. Fadin, *Inorg. Mater.: Appl. Res.*, **2017**, *8*, 32–36. DOI: 10.1134/s2075113317010063
179. T. T. Zhu, C. H. Zhou, F. B. Kabwe, Q. Q. Wu, C. S. Li, J. R. Zhang, *Appl. Clay Sci.*, **2019**, *169*, 48–66. DOI: 10.1016/j.clay.2018.12.006
180. Q. Yu, L. Zhu, T. Liu, S. S. Babu, Z. Zheng, S. Liu, Z. Chi, Y. Zhang, J. Xu, *Adv. Mater. Interfaces*, **2021**, *8*, 2001786. DOI: 10.1002/admi.202001786
181. Y.-Y. Yu, W.-C. Chien, T.-W. Tsai, *Polym. Test.*, **2010**, *29*, 33–40. DOI: 10.1016/j.polymertesting.2009.09.002
182. T. D. Mekuria, Z. Chunhong, L. Yingnan, D. El Din Fouad, K. Lv, M. Yang, Y. Zhou, *Mater. Chem. Phys.*, **2019**, *225*, 358–364. DOI: 10.1016/j.matchemphys.2018.12.107
183. Y. Lin, S. Hu, G. Wu, *J. Phys. Chem. C*, **2019**, *123*, 6616–6626. DOI: 10.1021/acs.jpcc.8b12519
184. M. Çakir, E. Akin, *J. Appl. Polym. Sci.*, **2019**, *136*, 47399. DOI: 10.1002/app.47399
185. M. J. Tommalieh, A. M. Zihlif, *Physica B: Condens. Mater.*, **2010**, *405*, 4750–4754. DOI: 10.1016/j.physb.2010.08.070
186. L. Zuo, W. Fan, Y. Zhang, L. Zhang, W. Gao, Y. Huang, T. Liu, *Compos. Sci. Technol.*, **2017**, *139*, 57–63. DOI: 10.1016/j.compscitech.2016.12.008
187. R. Magaraphan, W. Lilayuthalert, A. Sirivat, J. W. Schwank, *Compos. Sci. Technol.*, **2001**, *61*, 1253–1264. DOI: 10.1016/S0266-3538(01)00026-4
188. S. Chen, J. Xie, Z. Yang, *Polym. Bull.*, **2020**, *77*, 687–699. DOI: 10.1007/s00289-019-02767-2
189. K. H. Moon, B. Chae, K. S. Kim, S. W. Lee, Y. M. Jung, *Polymers*, **2019**, *11*, 489. DOI: 10.3390/polym11030489
190. Y. Mansoori, T. Mohseni Masooleh, *Polym. Compos.*, **2015**, *36*, 613–622. DOI: 10.1002/pc.22978
191. M. Dincă, J. R. Long, *Chem. Rev.*, **2020**, *120*, 8037–8038. DOI: 10.1021/acs.chemrev.0c00836
192. K. M. Jablonka, D. Ongari, S. M. Moosavi, B. Smit, *Chem. Rev.*, **2020**, *120*, 8066–8129. DOI: 10.1021/acs.chemrev.0c00004
193. J. L. Mancuso, A. M. Mroz, K. N. Le, C. H. Hendon, *Chem. Rev.*, **2020**, *120*, 8641–8715. DOI: 10.1021/acs.chemrev.0c00148
194. Z. Chen, H. Jiang, M. Li, M. O’Keeffe, M. Eddaoudi, *Chem. Rev.*, **2020**, *120*, 8039–8065. DOI: 10.1021/acs.chemrev.9b00648
195. A. J. Gosselin, C. A. Rowland, E. D. Bloch, *Chem. Rev.*, **2020**, *120*, 8987–9014. DOI: 10.1021/acs.chemrev.9b00803
196. M. D. Allendorf, R. Dong, X. Feng, S. Kaskel, D. Matoga, V. Stavila, *Chem. Rev.*, **2020**, *120*, 8581–8640. DOI: 10.1021/acs.chemrev.0c00033
197. Z. Zhang, H. T. H. Nguyen, S. A. Miller, S. M. Cohen, *Angew. Chem., Int. Ed.*, **2015**, *54*, 6152–6157. DOI: 10.1002/anie.201502733
198. Z. Zhang, H. T. H. Nguyen, S. A. Miller, A. M. Ploskonka, J. B. DeCoste, S. M. Cohen, *J. Am. Chem. Soc.*, **2016**, *138*, 920–925. DOI: 10.1021/jacs.5b11034
199. W. Li, *Prog. Mater. Sci.*, **2019**, *100*, 21–63. DOI: 10.1016/j.pmatsci.2018.09.003
200. Q. Qian, P. A. Asinger, M. J. Lee, G. Han, K. Mizrahi Rodriguez, S. Lin, F. M. Benedetti, A. X. Wu, W. S. Chi, Z. P. Smith, *Chem. Rev.*, **2020**, *120*, 8161–8266. DOI: 10.1021/acs.chemrev.0c00119
201. A. Bavykina, N. Kolobov, I. S. Khan, J. A. Bau, A. Ramirez, J. Gascon, *Chem. Rev.*, **2020**, *120*, 8468–8535. DOI: 10.1021/acs.chemrev.9b00685
202. T. Islamoglu, Z. Chen, M. C. Wasson, C. T. Buru, K. O. Kirlikovali, U. Afrin, M. R. Mian, O. K. Farha, *Chem. Rev.*, **2020**, *120*, 8130–8160. DOI: 10.1021/acs.chemrev.9b00828
203. S. Rojas, P. Horcajada, *Chem. Rev.*, **2020**, *120*, 8378–8415. DOI: 10.1021/acs.chemrev.9b00797
204. X. Liu, X. Wang, F. Kapteijn, *Chem. Rev.*, **2020**, *120*, 8303–8377. DOI: 10.1021/acs.chemrev.9b00746
205. A. M. Rice, C. R. Martin, V. A. Galitskiy, A. A. Berseneva, G. A. Leith, N. B. Shustova, *Chem. Rev.*, **2020**, *120*, 8790–8813. DOI: 10.1021/acs.chemrev.9b00350
206. L. S. Xie, G. Skorupskii, M. Dincă, *Chem. Rev.*, **2020**, *120*, 8536–8580. DOI: 10.1021/acs.chemrev.9b00766
207. D.-W. Lim, H. Kitagawa, *Chem. Rev.*, **2020**, *120*, 8416–8467. DOI: 10.1021/acs.chemrev.9b00842
208. A. E. Thorarindottir, T. D. Harris, *Chem. Rev.*, **2020**, *120*, 8716–8789. DOI: 10.1021/acs.chemrev.9b00666
209. M. J. C. Ordóñez, K. J. Balkus Jr, J. P. Ferraris, I. H. Musselman, *J. Membr. Sci.*, **2010**, *361*, 28–37. DOI: 10.1016/j.memsci.2010.06.017
210. T.-H. Bae, J. S. Lee, W. Qiu, W. J. Koros, C. W. Jones, S. Nair, *Angew. Chem., Int. Ed.*, **2010**, *49*, 9863–9866. DOI: 10.1002/anie.201006141
211. S. Sorribas, A. Kudasheva, E. Almendro, B. Zornoza, Ó. de la Iglesia, C. Téllez, J. Coronas, *Chem. Eng. Sci.*, **2015**, *124*, 37–44. DOI: 10.1016/j.ces.2014.07.046
212. T. Rodenas, I. Luz, G. Prieto, B. Seoane, H. Miro, A. Corma, F. Kapteijn, F. X. Llabrés I Xamena, J. Gascon, *Nat. Mater.*, **2015**, *14*, 48–55. DOI: 10.1038/nmat4113
213. M. Etxeberria-Benavides, O. David, T. Johnson, M. M. Łozińska, A. Orsi, P. A. Wright, S. Mastel, R. Hillenbrand, F. Kapteijn, J. Gascon, *J. Membr. Sci.*, **2018**, *550*, 198–207. DOI: 10.1016/j.memsci.2017.12.033
214. C. Zhang, Y. Dai, J. R. Johnson, O. Karvan, W. J. Koros, *J. Membr. Sci.*, **2012**, *389*, 34–42. DOI: 10.1016/j.memsci.2011.10.003
215. X. Ma, R. J. Swaidan, Y. Wang, C.-e. Hsiung, Y. Han, I. Pinnau, *ACS Appl. Nano Mater.*, **2018**, *1*, 3541–3547. DOI: 10.1021/acsanm.8b00682
216. F. Cacho-Bailo, G. Caro, M. Etxeberria-Benavides, O. Karvan, C. Téllez, J. Coronas, *Chem. Commun.*, **2015**, *51*, 11283–11285. DOI: 10.1039/c5cc03937a
217. F. Xie, N. Zhang, L. Zhuo, P. Qin, S. Chen, Y. Wang, Z. Lu, *Composites, Part B*, **2019**, *168*, 406–412. DOI: 10.1016/j.compositesb.2019.03.048

