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A REVIEW ON RECENT PROGRESS OF R&D FOR HIGH-TEMPERATURE RESISTANT POLYMER DIELECTRICS AND THEIR APPLICATIONS IN ELECTRICAL AND ELECTRONIC INSULATION

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Abstract. High-temperature resistant polymer dielectrics (HTPDs) have been finding great applications in electrical and electronic insulating fields due to their excellent thermal stability at elevated temperatures, good dielectric properties over wide temperature and frequency ranges, good mechanical properties, and high environmental stability, etc. The latest progress of research and development on HTPDs has been reviewed in the present paper. The developing history, structural features, molecular design, and functionalization for typical aromatic or heteroaromatic HTPDs, including imide polymers [polyimide (PI), polyamideimide (PAI), and polyetherimide (PEI), polybenzimidazole (PBI), polybenzoxazole (PBO), polyetheretherketone (PEEK), and polyphenylquinoxaline (PPQ) have been summarized. Then, the applications of HTPDs in electrical and electronic insulating areas were introduced. The future developing trend for HTPDs was prospected at last.

1. INTRODUCTION

Dielectrics have been becoming one of the most important components for both of electrical and electronic insulating applications because they can protect the equipments or devices from a variety of destroying such as dielectric loss, shorting out by sparking, dielectric breakdown, and so on [1-4]. According to the chemical nature of dielectrics, they can be roughly divided into three categories: inorganic dielectrics (ceramic, mica, glass, etc.), organic dielectrics (polymer, mineral oil, paper, etc.), and inorganic-organic hybrid dielectrics (polymer nanodielectrics, etc.). According to the state of molecular aggregation, dielectrics can also be divided as gas dielectrics (sulfur hexafluoride, etc.), liquid dielectrics (capacitor oil, etc.), and solid dielectrics (polymer, ceramic, etc.). Among various dielectrics, solid polymer dielectrics have been paid increasing attentions in recent years due to their excellent combined properties desired by engineering applications, including good dielectric features, good processing characteristics, and flexible structure designability [5]. Polymer dielectrics can be further divided into several classes according to their thermal endurance indices. For example, clear relative thermal endurance index (RTE or RTI) for electrical insulating materials (EIM) and electrical insulating systems (EIS) have been rated by standard NEMA (National Electrical Manufacturers Association) classifications, IEC (International Electrotechnical Commission) and Chinese national standard (GB/T) according to their maximum allowable operating temperature, as shown in Table 1.

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Table 1. Thermal class rating for EIM and EIS.

(International Electrotechnical Commission, IEC 60216) Chinese national standard (GB/T 11021-2014)			NEMA standard (for magnet wire)	
RTE	thermal class	alphabet	thermal class	alphabet or number
$90 \leq T < 105$	90	Y	90	O
$105 \leq T < 120$	105	A	105	A
$120 \leq T < 130$	120	E	120	E
$130 \leq T < 155$	130	B	130	B
$155 \leq T < 180$	155	F	155	F
$180 \leq T < 200$	180	H	180	H
$200 \leq T < 220$	200	N	200	200 (K)
$220 \leq T < 250$	220	R	220	220 (M)
$T \geq 250$	250	-	240+	C
			250	250

Typical polymer dielectrics corresponding to every thermal class are illustrated in Fig. 1.

Conventional polymer dielectrics, such as polyethylene (PE) or polypropylene (PP) intrinsically possess excellent insulating properties. However, they are facing great challenges in advanced electrical and electronic insulation engineering due to their limited service temperatures (≤105 °C). In recent years, with the development of electrical and electronic insulating applications to the trends of high power and high current, small and thin profile, high density, and high integrity, the demands to the high-thermal-class polymer dielectrics has been increasingly emphasized so as to maintain high reliability of the devices operating at high temperatures [6,7]. High temperature is usually defined as an operating temperature higher than 175 °C [8]. For instance, for the recent aviation and aerospace electrical machine winding applications, the electrical insulating system must withstand the high temperature environment in excess of 200 °C with lifetimes of several thousands of hours for the engines [9]. Elevated temperature environment usually increases mechanical stresses because of coefficient of thermal expansion (CTE) mismatches among various materials in the systems, accelerates corrosion process, and degrades electrical properties for the dielectrics. High-temperature also reduces mechanical strength, increases creep, or even causes material phase transition or decomposition in the dielectrics. Additionally, dielectric leakage in electrical or electronic devices (such as semiconductor devices, capacitors, etc.) increases with temperature. One of the most important issues in designing high-temperature electrics and electronics is to select high performance dielectrics and struc-

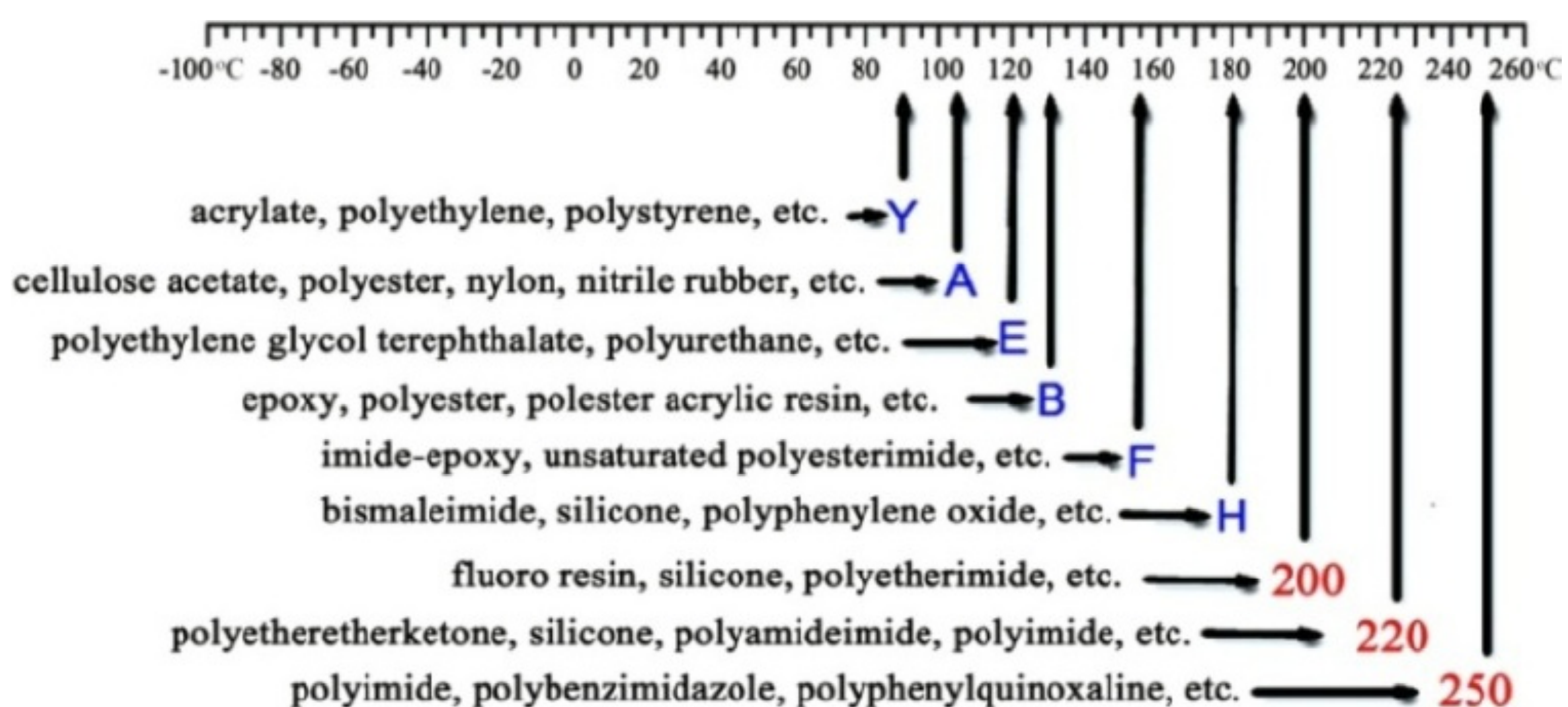

Fig. 1. Typical polymer dielectrics and their thermal endurance index values.

tures to minimize these failure modes. Apparently, conventional polymer dielectrics will lose their dielectric and mechanical properties at such high temperatures. Thus, high-temperature resistant polymer dielectrics (HTPDs) with thermal class over H (>180 °C) have attracted much attention from both of the academic and industrial engineering.

As shown in Table 1 and Fig. 1, HTPD materials usually include fluoro resin, silicone resin, aromatic polymers, such as polyetheretherketone (PEEK), and heteroaromatic polymers, such as polyimide (PI), polyamideimide (PAI), polyetherimide (PEI), polybenzimidazole (PBI), polybenzoxazole (PBO), polyphenylquinoxaline (PPQ), etc. The common structural characteristics for HTPDs include high-energy chemical bonds (C-F, Si-O, etc.) in the molecules, strong inter- and intra-molecular interactions caused by hydrogen bond (-NH in PBI or –CO-NH- in PAI, etc.), or conjugation action along the polymer chains (imide ring, imidiazole ring, etc.) [10]. These structural features often endow HTPDs with high thermal stability, excellent dielectric properties as well as good mechanical properties and environmental stability. Excellent combined properties make HTPDs good candidates for high-temperature insulating applications.

Considerable progress has been achieved in both of the academic development and commercialization for novel HTPD materials in the past decades. Since the R&D of common fluoro resin, silicone resin, and aromatic polyamide types of HTPDs has been well reviewed in the literatures [11-13], the state of art and future developing trends for rarely-reviewed hetero- or aromatic-ring HTPDs, including PI, PAI, PBI, PBO, PEEK, and PPQ in electrical and electronic insulation was introduced in the current work. The molecular design and performance characters for HTPD materials were presented. The applications of HTPDs in several important electrical and electronic fields were also reviewed.

2. HIGH TEMPERATURE RESISTANT POLYMER DIELECTRICS

2.1. Brief introduction for high temperature polymers

High temperature polymers (HTPs) are usually characterized by their superior physical and chemical property stability at elevated temperatures compared to common organic polymers [14]. Briefly, the development of HTPs has experienced three stages up to now. The first stage is the origin and initial mass-production for HTPs in 1960s, largely driven by the demands of human space exploration. Various HTPs, including, PI, PBI, PBO, and "ladder" polymers were designed and produced. However, a large proportion of the HTPs gradually died out in the following years due to poor processability or extremely high cost. The second developing stage for HTPs is in the range of 1970s to 1980s, with the rapid development of electrical and electronic industry, especially microelectronic industry. In this stage, for the HTPs originated in 1960s, only those possessing both of high thermal stability, good processability, and relatively low cost survived and achieved commercialization. The third stage is what we are experiencing now, that is the functionalization of HTPs for applications in advanced electrical (high-speed transportation system, high-power energy generation, storage and transmission, etc.) and electronic (microelectronic and optoelectronic fabrication) applications.

For understanding the applications of HTPs in electrical and electronic insulation, we can find the trace from the relationship between the years synthetic polymers started being applied to insulation and their highest usable temperatures [15]. From the mid-1960s, HTPs have been increasingly adopted as dielectrics in insulation applications. This developing trend is closely related with the urgent demands of high-temperature and high-reliability electrics and electronics. Various applications of HTPs in electrics and electronics have been reviewed in the literature during the past decades [16].

Generally, most of the demands on the properties of HTPDs for electrical and electronic applications are similar, including high thermal stability, high dielectric strength, high surface and volume resistivity, and so on. However, there still have different focuses for these two applications. Thus, when designing HTPDs for the specific applications, one usually addresses different molecular design emphasis. Fig. 2 summarized various molecular design procedures, including favorable designs and unfavorable ones in developing HTPDs for electrical and electronic applications. The unfavorable procedures for these two aspects are usually the same, both of which are in order to preventing the deterioration of thermal resistances for the HTPDs. However, the favorite ones are somewhat different. For instance, for some specific electrical applications, such as embedded capacitors, they usually demand HTPD materials with a high dielectric constant at high frequency, a low leakage current, and a reasonable high breakdown field [17-20]. Thus, polar and planar substituents, such as nitrile group [21]

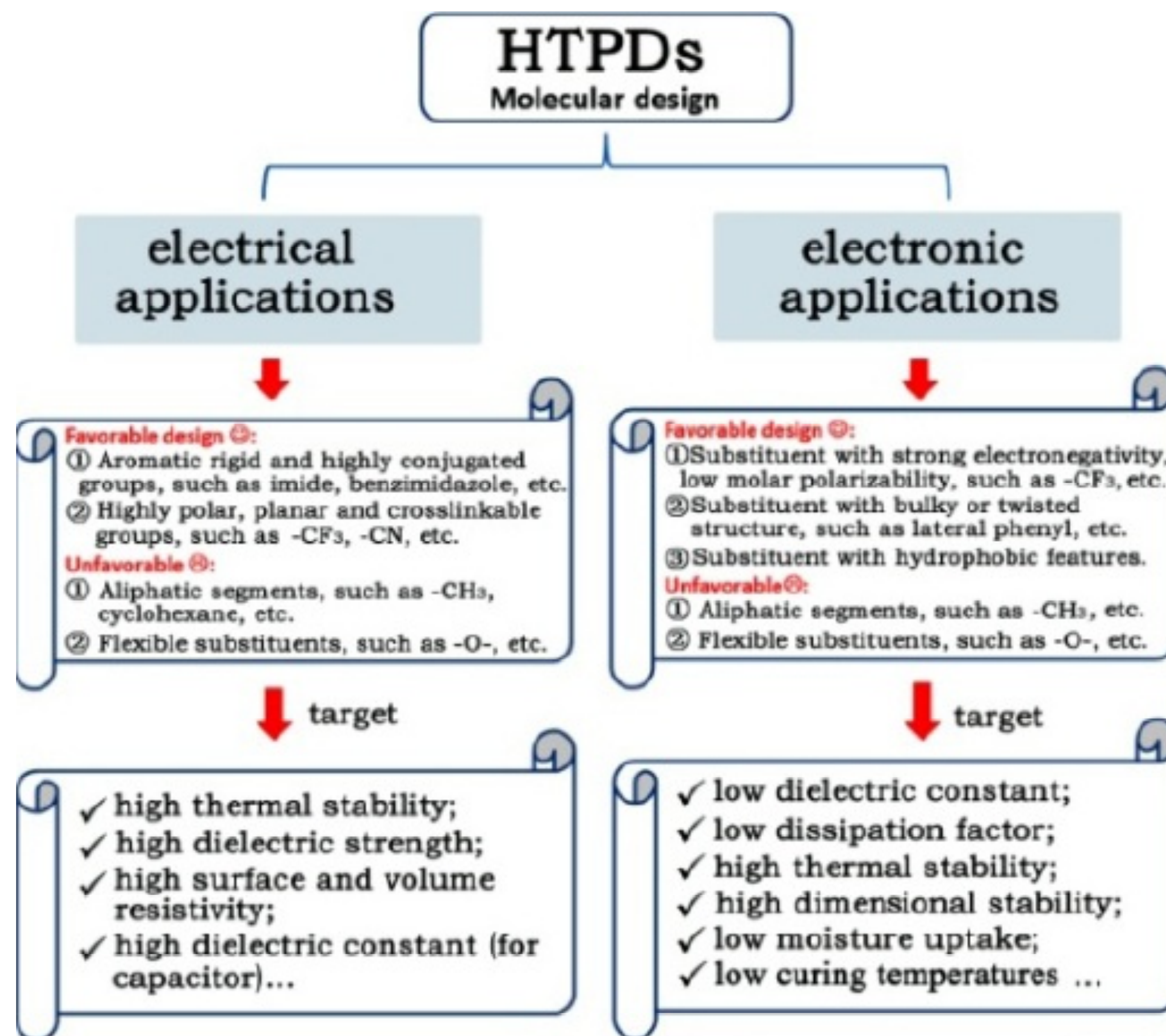

Fig. 2. Property requirements of electrical and electronic applications for HTPDs.

and short-chain alkyl groups [22] are favorable in the design because they can increase the dielectric constants of the dielectrics. However, for electronic applications, HTPDs with low dielectric constants (low-*k*) and dissipation factors are highly desired because they can lower line-to-line noise in interconnects and alleviate power dissipation issues by reducing the capacitance between the interconnect conductor lines. In addition to providing device speed improvements, low-*k* interlayer dielectrics also provide lower resistance-capacitance delay, making them superior to low resistivity metal conductors such as copper and silver [23-25]. At this point, HTPDs with apolar and twisted or bulky molecular structures are preferred. In addition, for microelectronic applications, HTPDs with relatively lower curing temperatures (<250 °C) are preferable because many kinds of microelectronic components are temperature sensitive [26].

2.2. High temperature resistant polymer dielectrics

As mentioned above, HTPDs have been achieving rapid development both in the basic research and commercialization in the past decades. Fig. 3 summarizes the typical commercially available HTPDs. Their typical chemical structures and common preparation procedures are illustrated in Fig. 4 [27]. Various forms, including varnish, coating, adhesive, sealant, film, fiber, plastics, etc. have been developed for HTPDs according to different applications. Generally, liquid HTPD forms are suitable to be used in wires, cables, and integrated circuits applications; whereas solid ones in printing circuit boards and various insulating connectors.

All of these HTPDs possess aromatic or heteroaromatic molecular skeletons in order to maintain high thermal, dielectric, and dimensional stability at elevated temperatures. Some easily thermal-degradable groups, such as flexible ether linkages (for PEI, PEEK, etc.) or lateral phenyl substituent (for PPQ) have also to be introduced in order to achieve good processability, such as direct heat-sealability to copper wires without using any additional adhesives. For the rigid HTPDs with insoluble and infusible characteristics, such as PI and PBO, their easily-processable precursors are often used in practical applications [28]. The precursors will turn into the final HTPDs by curing at elevated temperatures. This methodology is widely used in electrical and electronic insulation applications.

The typical thermal and dielectric properties for the HTPDs can be clearly understood from the data reported by the supplier or shown in the literatures, which are briefly tabulated in Table 2. Obviously, high glass transition temperatures (T_g), high resistivity, and high dielectric strength are the common features for the HTPDs. The specific characteris-

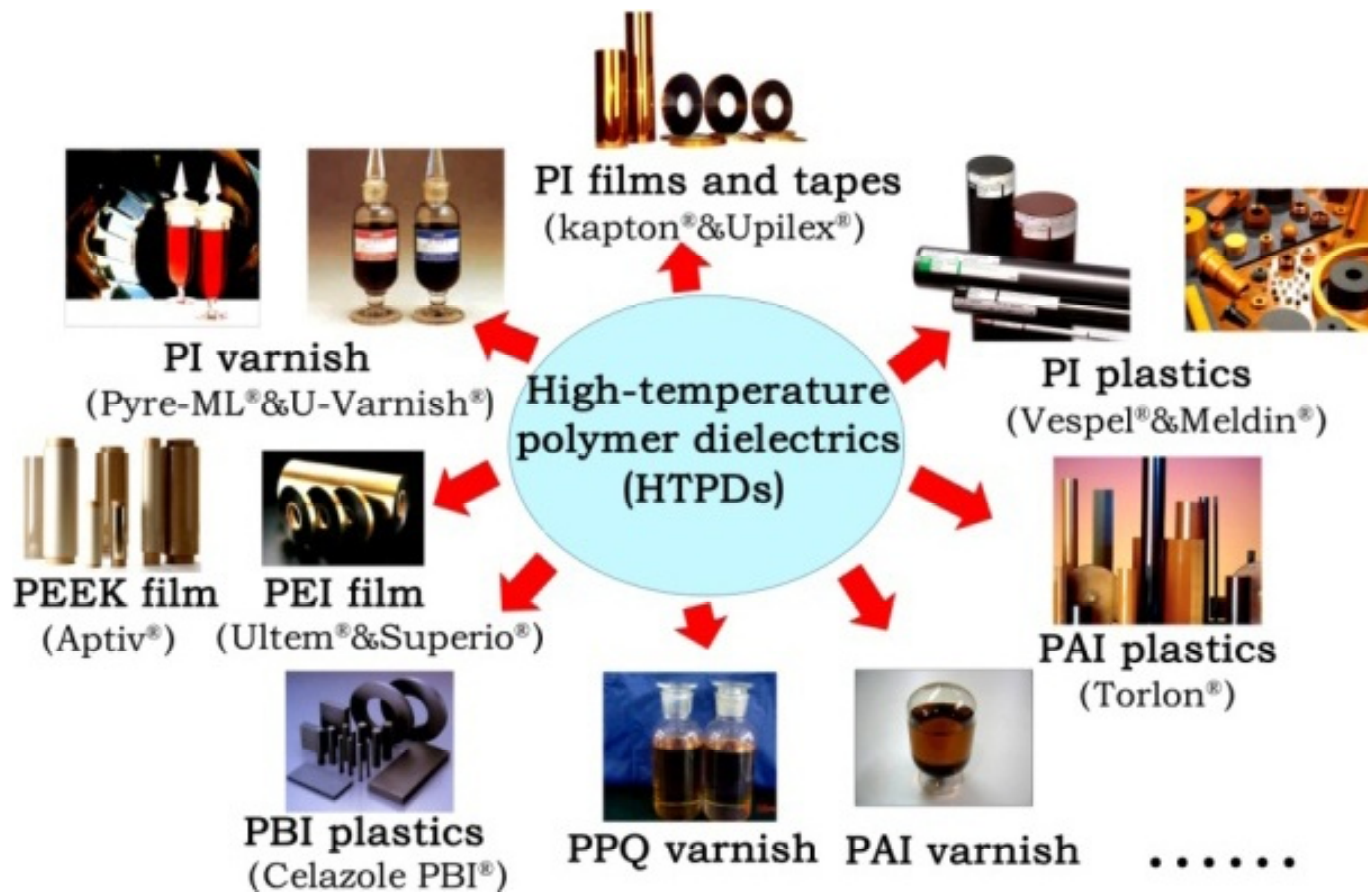

Fig. 3. Representative commercially available HTPDs in insulation applications.

Fig. 4. Common preparation procedures for HTPDs.

Table 2. Typical thermal and dielectric properties for common HTPDs.

HTPDs*	T_g(°C)	Volume resistivity (Ω cm)	Dielectric strength (kV/mm)	Dielectric constant	Dissipation loss factor
PI film Kapton® HN	>300	1.5×10^{17}	303 (25 um)	3.4 (1 kHz)	0.002 (1 kHz)
PI film Kapton® CRC	>300	2.3×10^{16}	291 (25 um)	3.9 (1 MHz)	0.003 (1 MHz)
PI film Kapton® MT	>300	$>10^{16}$	220 (25 um)	4.2 (1 MHz)	Not available
PI film Upilex®-S	>350	10^{15}	272 (25 um)	3.5 (1 kHz)	0.001 (1 kHz)
PI plastics Vespel®-SP1	>300	10^{14}-10^{15}	22.0 (2 mm)	3.5 (1 MHz)	0.003 (1 MHz)
PI plastics Meldin® 7001	>300	10^{15}-10^{16}	22.9 (2 mm)	3.1 (1 MHz)	Not available
PAI plastics Torlon®4203L	>300	2.0×10^{17}	23.6 (2 mm)	3.9 (1 MHz)	0.031 (1 MHz)
PBI plastics Celazole® PBI	427	2.0×10^{15}	23.0 (2 mm)	3.2 (1 MHz)	0.003 (1 MHz)
PEI plastics Ultem®-1000	219	2.1×10^{17}	200 (25 um)	3.2 (1 MHz)	0.005 (1 MHz)
PEI plastics Extem® XH1005	267	$>10^{16}$	25 (1.5 mm)	3.1 (1 MHz)	0.092 (1 MHz)
PBO varnish HD-8820	300	3.0×10^{16}	470 (3 um)	2.9 (1 MHz)	0.009 (1 MHz)
PEEK film Aptiv® 1000	Not available	Not available	190 (50 um)	3.5 (10 MHz)	0.002 (10 MHz)
PPQ film IP200®	365	3.0×10^{17}	-	2.4 (1kHz)	0.0003 (1kHz)

* Katpon® HN: poly(pyromellitic dianhydride-co-4,4 -oxydianiline), common PI film Kapton® CR100: corona-resistant PI film; Kapton® MT100: thermally conductive PI films, Dupont, USA; Upilex®-25S: poly(3,3',4,4'-biphenyltetracarboxylic dianhydride-co-1,4-phenylenediamine), high-temperature PI films, Ube, Japan; Vespel®-SP1: unfilled PI engineering plastics, Dupont, USA; Meldin® 7001: unfilled PI engineering plastics, Saint-Gobain, France; Torlon®4203L: unfilled PAI engineering plastics, Solvay, Belgium; Celazole® PBI U-60: unfilled PBI engineering plastics, Celanese, USA; Ultem®1000: common PEI film, Extem® XH1005: high temperature PEI plastics, Sabic, Saudi Arabia; HD-8820: photosensitive PBO varnish, HD-Microsystems, Japan; Aptiv® 1000: common PEEK film, Victrex, England; IP-200®: common PPQ film, IFP-Cemota, France.

tics for every HTPD will be briefly introduced and compared as follows.

2.2.1. Imide polymers (Polyimide, polyamideimide, polyetherimide)

Imide polymers are characterized by the five-membered imide ring in the backbone of the polymers [29]. The physical and chemical characteristics of the imide cyclic structure determine the basic properties for the polymers, including favorable high thermal and thermal-oxidative stability, high dielectric strength, high mechanical strength, and unfavorable moisture sensitive and high water uptakes.

(1) Polyimide (PI). PI is one of the most widely used HTPDs in both of electrical and electronic applications. Meanwhile, PI is the most commercialized species in HTPD families. Various PI forms, including PI impregnating varnish (usually PI precursors), PI coatings, PI films, and PI engineering plastics have been successfully used in insulation industry for half a century. The typical preparation procedure for PI is shown in Fig. 4. Generally, PI varnishes or enamels with the form of their soluble precursors poly(amic acid) (PAA) in polar solvents, such as *N,N*-dimethylacetamide (DMAc) are widely used for high-temperature magnet wire insulation [30]. During the high temperature baking, the solvents are evaporated, and at the same time, the PAA precursors are dehydrated to facilitate the ring closure with the elimination of water to afford the final PI coatings with a thermal class over 220 °C. This chemical reaction is known as imidization reaction, which usually needs the high temperature as high as 350 °C to be complete. In order to avoid the pin-holes and other defects in the PI coatings, a step-by-step procedure is usually adopted for the imidization and

PAA precursors. Some representative PI dielectrics are introduced as follows.

First, general-purpose PI films (such as Kapton® HN film by Dupont, USA with the chemical structure shown in Fig. 4). These PI films usually provide an excellent balance of properties over a wide range of temperatures (-269-400 °C). The films with a thickness of 25 μm show typical dielectric properties of a thermal class of 220 – 240 °C, dielectric strength around 300 kV/μm, volume resistivity over 10^{17} Ω cm, dielectric constant of 3.2-3.4 at 1 kHz, and dissipation factor lower than 0.002 at 1 kHz. In addition, the film has excellent thermal stability with T_g values higher than 350 °C. Secondly, high temperature resistant PI films (such as Upilex®-S PI film by Ube, Japan). Upilex®-S film is a super-heat resistant PI film deriving from biphenyl-type monomers, which is different from that of Kapton® HN (Fig. 4). This formulation makes the PI film exhibit outstanding thermal and thermo-oxidative stability, dimensional stability, excellent dielectric properties, low water absorption and very high chemical resistance. Tsukiji and coworkers evaluated the thermal degradation of endurance of Kapton® HN and Upilex®-S films in severe environments [31]. The experimental results revealed that both of the two PI films were able to be used in electrical insulation systems operating at temperatures up to 350 °C for 20000 hours in inert gas. In air, only Upilex®-S was able to be used at 300 °C. The diphenylether moiety in Kapton® HN film was thought to be vulnerable to thermal aging in air. For insulating applications, Upilex®-S film shows excellent electrical characteristics (thermal class: 250 °C; dielectric strength: 272 V/μm for 25 μm film and 440 kV/μm for 75 μm film at both of 25 °C and 200 °C) over a wide range of temperatures and frequencies. Even at high temperatures, the film shows almost no deterioration in its electrical properties. Diaham and coworkers investigated the dependence of dielectric breakdown of Upilex®-S film on the area, thickness, and temperature [32]. In the test temperature range of 25-400 °C under direct circuit (DC) ramps, no temperature-dependence of the dielectric strength was observed. The dielectric breakdown field of the film remained relatively high (>2 MV/cm) up to 400 °C. Upilex®-S film also exhibits a low level of insulating defects, making it an optimal choice for electrical and electronic applications that demand high reliability. The other features for the film include small values for both heat shrinkage and thermal linear expansion coefficients (CTE: 12 ppm/°C from 50-200 °C) and high flame retardancy (UL94 VTM-0). These features make the film ideal for use in high-temperature electronic insulating application. Thirdly, corona-resistant PI films (such as Kapton® CRC film by Dupont, USA). These films are developed specifically to withstand the damaging effects of corona, which can cause ionization and eventual breakdown of insulating dielectrics when the voltage stress reaches a critical level. Kapton® CRC PI film shows corona resistance or voltage endurance greater than 100,000 h at 20 kV/mm at 50 Hz. The film also provides twice the thermal conductivity (0.385 W/m K) of standard PI film. These combined properties make Kapton® CRC film good candidate for applications in the insulation of transformers, traction motors, frequency conversion motor and electrical rotating machines. Fourthly, thermal conductive PI films (such as Kapton® MT film by Dupont, USA, etc.). The PI film usually offers an excellent combination of electrical properties, thermal conductivity and mechanical properties, making it ideal for use in controlling and managing heat in electronic insulating applications, such as printed circuit boards, etc. Fifthly, PI engineering plastics (such as Vespel® SP1 by Dupont, USA; Meldin® 7001 by Saint-Gobain, France). The PI engineering plastics are usually processed into various parts and shapes for electrical and electronic insulation applications. The parts are usually more ductile than ceramics and light than metals. They have outstanding dielectric properties over wide temperature (25-250 °C) and frequency (10^2-10^5 Hz) ranges. The combination of good dielectric properties, high strength and excellent thermal and radiation resistance makes the PI plastics outstanding candidates for insulations in severe environments.

Although PIs have found various applications in electrical and electronic insulation, the insoluble and infusible nature limited their applications. For example, in magnet wire insulation, PI films have to be combined with heat-fusible fluoropolymer film, such as fluorinated ethylene propylene (FEP) film in order to achieve hot windings. This methodology has been well established for various PI dielectrics, such as heat-sealable common PI films (Kapton® FN), heat-sealable corona resistant PI films (Kapton® FCRC), and so on. Alternatively, other imide polymers with enhanced thermo-processability, such as PAI and PEI have also been developed.

(2) Polyamideimide (PAI). PAI combines the good property merits of PI and wholly aromatic polyamide. Thus, the polymers usually possess good thermal stability, excellent dielectric and mechanical properties, good processability, and chemical resistance. PAI can usually be used with two formats - varnish and plastics in insulation industry. As shown

in Fig. 4., from the aspect of property or cost, the PAI resin used most often for enameled wires can be obtained mainly by a synthesis reaction of 4,42 -diphenylmethane diisocyanate (MDI) and trimellitic anhydride (TMA) in a polar, aprotic solvent such as *N*-methyl-2-pyrrolidinone (NMP) at high temperatures. The obtained PAI resins are soluble in solvents with the fully cyclized forms to afford the PAI enamels [33].

PAI varnish are widely used as class 220 °C enamels and applied mostly as overcoat to produce dual coating wires using for hermetic motors and Freon-resistant wires [34]. In addition, with the tremendous speed increase for winding of motors and other electrical devices, PAI have been widely used as magnet wire coatings due to the good abrasion resistance and self-lubricating feature in order to maintain low defect rates for the products. Typically, in electrical insulations, the wire enamel structure of polyesterimide (PEsI) layer underneath and PAI on top of it is the most common magnetic wires used in low-voltage machines operated under high temperature. This insulated magnetic wire has very good mechanical properties, making the winding process easier because the wire can be stretched and bent roughly. PAI engineering plastic (such as Torlon® 4203L by Solvay, Belgium, etc.) has high strength and stiffness up to 260 °C and can be processed into various insulating parts, even with complicated structures via the low-cost injection molding procedure. This characteristic greatly expands the applications of PAI in insulation industry.

(3) Polyetherimide (PEI). PEI represents a modified PI with flexible ether linkages in the repeating unit of the polymer. A typical of preparation procedure for PEI is shown in Fig. 4. Due to its high solubility in solvents, PEI can usually be prepared by the one-step high-temperature polycondensation procedure using bisphenol A dianhydride (BPADA) and *m*-phenylenediamine as the starting monomers. Although introduction of ether groups inevitably sacrifices the thermal stability of the pristine PI, good processability, such as heat sealability is usually achieved. PEI dielectrics are heat sealable to a variety of materials, including metal (copper foils, etc.) and other polymers (PI, etc.) due to their thermoplastic nature; thus can be wound to the wires without using any additional adhesives. In addition, PEI exhibits good resistance to ultraviolet radiation and gamma radiation without stabilizers. Thus it might be used in nuclear radiation environments for long terms [35].

PEI can be available with the forms of film and engineering plastics. The first generation of PEI, such as Ultem® 1000 (Sabic, Saudi Arabia) shows a T_g value of 219 °C, which can usually meets the class H electrical insulation requirements for motor and transformer applications. The good design flexibility for PEI polymers has now offered the second generation of high-T_g type alternative PEIs, such as Extem® XH1005 (T_g: 267 °C by Sabic, Saudi Arabia), which can meet the demands of class 200 insulation. Very recently, PEI resin (Ultem® 9085, Sabic, Saudi Arabia) which can be processed into various insulating parts by 3D printing technique has also been reported [36].

2.2.2. Polybenzimidazole (PBI) and polybenzoxazole (PBO)

PBI and PBO are initially commercialized as high-performance fibers due to their high strength, high modulus, and excellent flame retardancy [37]. As illustrated in Fig. 4, conventional PBI and PBO polymers are usually prepared via high-temperature polycondensation procedures so as to achieve high degree of cyclization. Lately, it was discovered that these polymers have excellent properties as dielectrics for insulation applications. Thus, various insulating varnishes, coatings, enamels and plastics based on these polymers have been developed. For example, PBI plastics (such as Celazole® PBI U-60 by PBI Performance Products, USA) are known as the highest thermal stable thermoplastics with a T_g higher than 400 °C. Electrically insulating parts made from PBI have fine electrical properties, good chemical stability, flame retardancy, and excellent resistance to heat, cold, radiation, and other severe environments. Recently, PBI coatings (such as S10 and S26 solution by PBI Performance Products, USA) with solid contents higher than 10 wt.% in polar solvent achieve commercialization, which are suitable for film casting, dip coating, spray coating and resin impregnation. The cured coating has the similar properties with PBI matrix. These products have great potential applications in high-temperature magnet wire insulation.

PBO has the advantage over other high-temperature polymer films due to the absence of polar groups in its repeating unit. The absence of polar groups in the polymer prevents the formation of hydrogen bond between the polymer and water, therefore resulting in less water absorption in the film and low dielectric constant (<3.0 at 1MHz) and dielectric loss factor. Additionally, PBO is known for its excellent thermal stability and thermo-oxidative resistance, high electrical resistivity, and high dielectric strength that remains stable over a long temperature period. Re-

cently, a key technique achieving fully cyclization of PBO at temperatures lower than 250 °C has been achieved [38]. This curing temperature is much lower than that of common PI dielectrics which have to be cured at 350 °C or higher. A series of low-temperature curable PBO dielectrics have been commercialized recently by HD-Microsystems, Japan, including HD-8921 with curing condition of 250 °C for 1 h; HD-8930 of 225 °C for 1 h, and HD-8940 of 200 °C for 1 h. This feature is critical for most microelectronic and optoelectronic applications, in which many components are high temperature sensitive. The good combined properties make PBO good candidates for in microelectronic packaging.

2.2.3. Polyphenylquinoxaline (PPQ)

PPQ represents a class of high performance thermoplastic heteroaromatic polymers, which is characterized by the phenyl-substituted quinoxaline ring in the structure [39]. The special molecular structure endows the polymers many favorable properties, such as high thermal and thermo-oxidative stability; good solubility in organic solvents and relatively low dielectric constants; and excellent hydrolytic resistance. Thus, PPQs have found various potential applications as dielectrics for electrical and electronic fabrication [40]. Although PPQs have been extensively studied for half a century, they have not been achieved wide commercialization like the other heteroaromatic polymers, such as PI. This is mainly due to the relatively high cost and very limited commercial availability of the starting monomers, including bis(α-diketone) and bis(o-diamine) compounds shown in Fig. 4. Nevertheless, PPQ still attracts the attention in dielectric materials R&D because the polymer has been proven to be one of the best hydrolytic resistant high temperature polymers and possesses excellent hydrolysis resistance to various (neutral, acidic or alkaline) waters not only at room temperature, but at elevated temperatures. PPQ films could maintain integral shapes and flexibility even after boiling in distilled water at 250 °C (vapor pressure of water: 3.9 MPa) for 60 h. Under the same conditions, the PI films have been completely hydrolyzed. Commercially available IP200® PPQ films are not degraded when aged for one month in 50% sodium hydroxide solution up to 150 °C or 40% sodium hydroxide solution up to 160 °C. They are also stable in boiling water up to one year. This unique characteristic makes PPQ good candidates in various insulation applications with high humidity and high temperature circumstances. In addition, the bulky phenyl-substituted quinoxaline rings in PPQs is nonpolar, so the dielectric constants of PPQs are usually lower than those of other high temperature polymers. This feature makes PPQs a good low dielectric constant (low-*k*) material as the interlayer dielectric (ILD) for ultra-large integrated circuit (ULSI) fabrications [41]. Recently, Liu and coworkers developed a low-cost route for PPQs. Various functional PPQs with relatively low cost while maintaining intrinsic thermal stability, dielectric properties, and hydrophobic nature have been developed [42-45]. This endeavor greatly expands the wide application of PPQ in insulating industry.

2.2.4. Polyetheretherketone (PEEK)

PEEKs are a family of polymers containing alternative ether and ketone groups in their repeating units, whose typical synthesis procedure is shown in Fig. 4. Such kind of chemical structure imparts PEEKs semi-crystalline nature with high thermal resistance and excellent dielectric properties. In addition, unlike the other heteroaromatic polymers mentioned above, PEEK can usually be winded with copper or aluminum wires directly at elevated temperature without using any adhesives. This is quite beneficial for improving the reliability of the insulating products. At last, PEEK usually possesses very stable dielectric properties over a wide range of temperatures, frequencies and humidity. Especially, the good resistance to steam makes PEEK unique dielectric for insulating applications because steam is a very harsh environment for insulating materials. It is a combination of water and high temperature and easily causes the hydrolysis of common HTPDs. It has been proven that PEEK can resist hydrolysis in temperatures up to 250 °C [46].

PEEK can be used as HTPDs with both forms of engineering plastics and film. The recently commercialized PEEK film, Aptiv® 1000 (Victrex, England) has a semi-crystalline molecular structure, exhibiting superior moisture, chemical, abrasion, and radiation resistance to other polymer dielectric films. The melt-processable PEEK film has a thermal index of 220 °C.

3. ENGINEERING APPLICATIONS OF HTPDS

3.1. Electrical insulation

3.1.1. High-temperature magnet wire insulation

Enamel-insulated copper and aluminum magnet wires are used principally in the electrical and elec-

tronics industries for coils, inductors, transformers, generators, armatures, solenoids, and other windings [47]. Wire may be insulated and protected with a wide variety of coating materials which may be classified according to the manner of application. For example, wires may be dipped in a liquid bath of the resin solution (known as varnish, enamel or impregnating coating, etc.) or coated by extrusion of solid thermoplastic resins. For HTPDs, they can usually be applied to the wires either by dip coating or by extruded coating, although the former manner is used more frequently.

As mentioned above, magnet wire may be classified into several ratings according to NEMA standard (Table 1). For high temperature magnet wires with the maximum serving temperature over 200 °C, thermal class is clearly the most important parameter to be considered when designing the dielectrics. For instance, Petitgas, et al. investigated the relationships between chemical structure and electrical behavior in the high temperature aging of copper wires enameled with various HTPDs, including crosslinked polyesterimide (PEsI) with a thermal class of 180-200 °C, PAI with a thermal class of 220 °C, and PI (PAA precursors) with a thermal class of 240 °C[48]. Copper wires were enameled by PEsI (thickness: 0.025 mm), PEsI+PAI (thickness: 0.035 mm), PAI (thickness: 0.03 mm), and PI (thickness: 0.035 mm), respectively. All the wire samples were thermally aging in the temperature range of 200-400 °C in an oven. The experimental results revealed that the enameled wires based on PEsI started to degrade above 300 °C, whereas those based on PAI and PI were stable before 400 °C. Meanwhile, PAI presented promising performances for high temperature rotating machine applications compared to those of PI. Schadler and coworkers investigated the high temperature breakdown strength and voltage endurance of nanofilled PAI dielectrics [49]. In the experiments, nanoscale silica and alumina were filled into PAI, respectively, for high temperature wire enamel applications. Experimental results showed that, at filler loading of 5-7.5 wt.%, the direct circuit (DC) breakdown strength improved for both the PAI systems at 30 °C and 300 °C; however, only the alumina system exhibited an increase in alternative circuit (AC) breakdown strength. The authors ascribed this phenomenon to the electron scattering and corona resistance of the alumina nanoparticles. This result is quite beneficial for designing HTPDs with improved dielectric strength. Recently, researchers from Kennedy Space Center (KSC), National Aeronautics and Space Administration (NASA) reported a PI wire insulation repair system [50]. In the technique, thin film PI patches are applied to the damage areas of wire insulation with a heating device that adhering the PI repair film into the place. The wire repairs made with the system are permanent, flexible and much less intrusive than repairs made using current techniques and materials. This technology is well suited for all applications of PI and other high temperature polymer enameled wire constructions and might find potential applications in aerospace wiring, automotive wiring, and other industrial fields. The used patches are low-temperature meltable PIs derived from bisphenol A dianhydride and aliphatic or aromatic diamines and double bonds-containing crosslinker. The cured PI patches have good electrical properties.

For industrial applications, various magnetic wires enameled with HTPDs have been achieving wide applications in high-tech areas. For instance, PI insulated wires and cables, such as ESCC 3901 001 developed by Axon2 Cable&Interconnect, France with the operating temperature of -100~200 °C have been successfully applied in the electrical systems for various space projects, including LISA (Laser Interferometer Space Antenna) Pathfinder for low-frequency gravitational wave detection [51], Eurostar 3000 satellite platform [52], and so on. The similar wires and cables insulated with PI/ fluorothermoplastic (ESCC 3901 018) with the operating temperature of -200~200 °C have been applied in the electrical system of GOCE (gravity field and steady-state ocean circulation explorer) spacecraft operating in low orbit orbits [53]. Excellent thermal, dielectric and radiation stability for the PI coatings enable them good insulating protection for the spacecrafts.

3.1.2. Aerospace and aviation electrical machines insulation

HTPDs have been evaluated as dielectric films for aviation and aerospace electrical insulation for several decades. For example, PBI, together with poly-P-xylene (PPX) and Teflon® perfluoroalkoxy (PFA) were characterized to explore their possible use as a replacement for PI (Kapton®) in aerospace high voltage wiring insulation [54]. Although PI (Kapton®) has been widely used for wire insulation and energy storage for aerospace and space power systems, some performance defects including arc-tracking and cracking under a combination of high temperature and humidity limited its reliability. The evaluation results indicated that PBI exhibited excellent thermal stability and dielectric strength in the temperature region of 250-300 °C as compared to

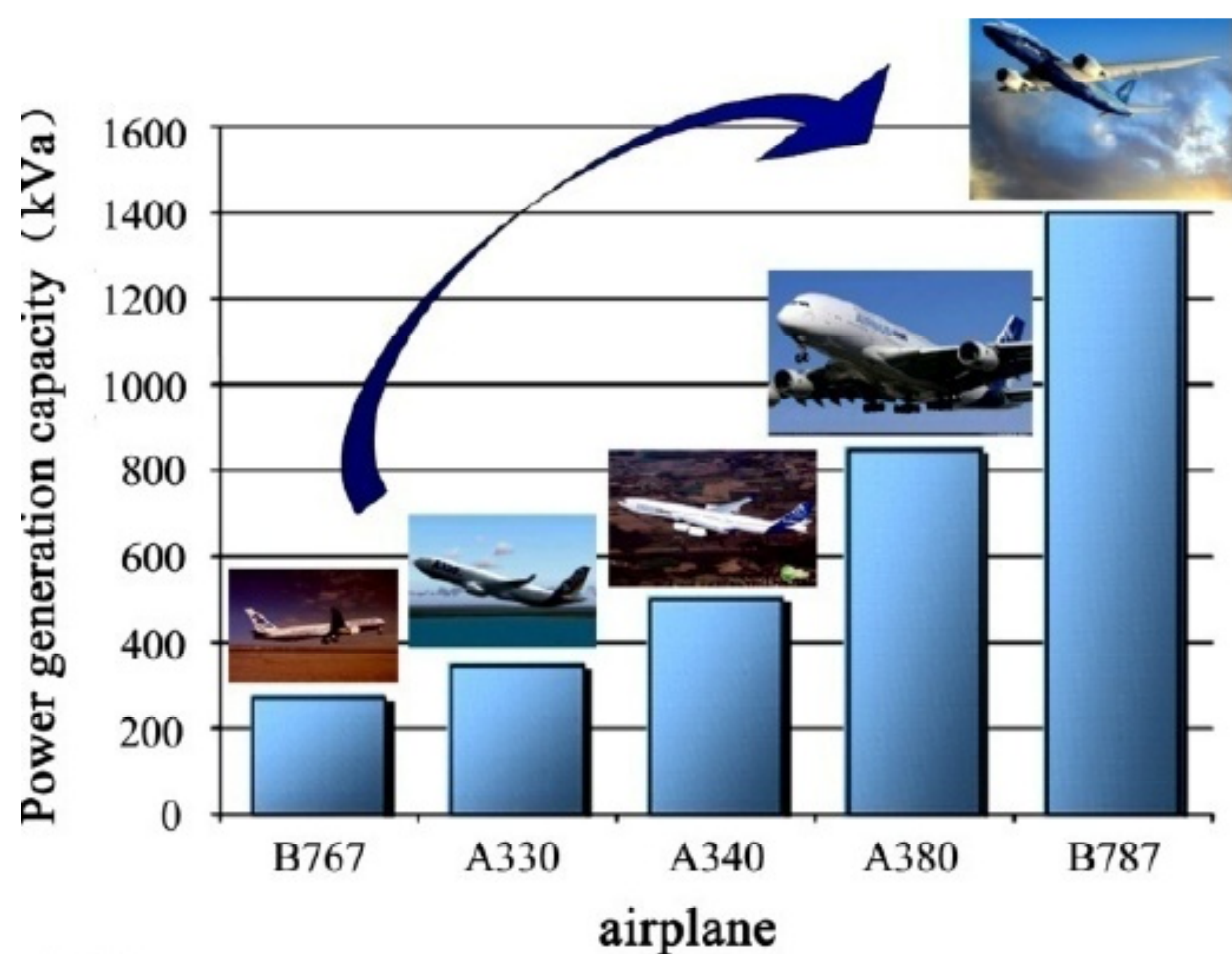

Fig. 5. Aircraft electrical power generating capacity by aircraft type.

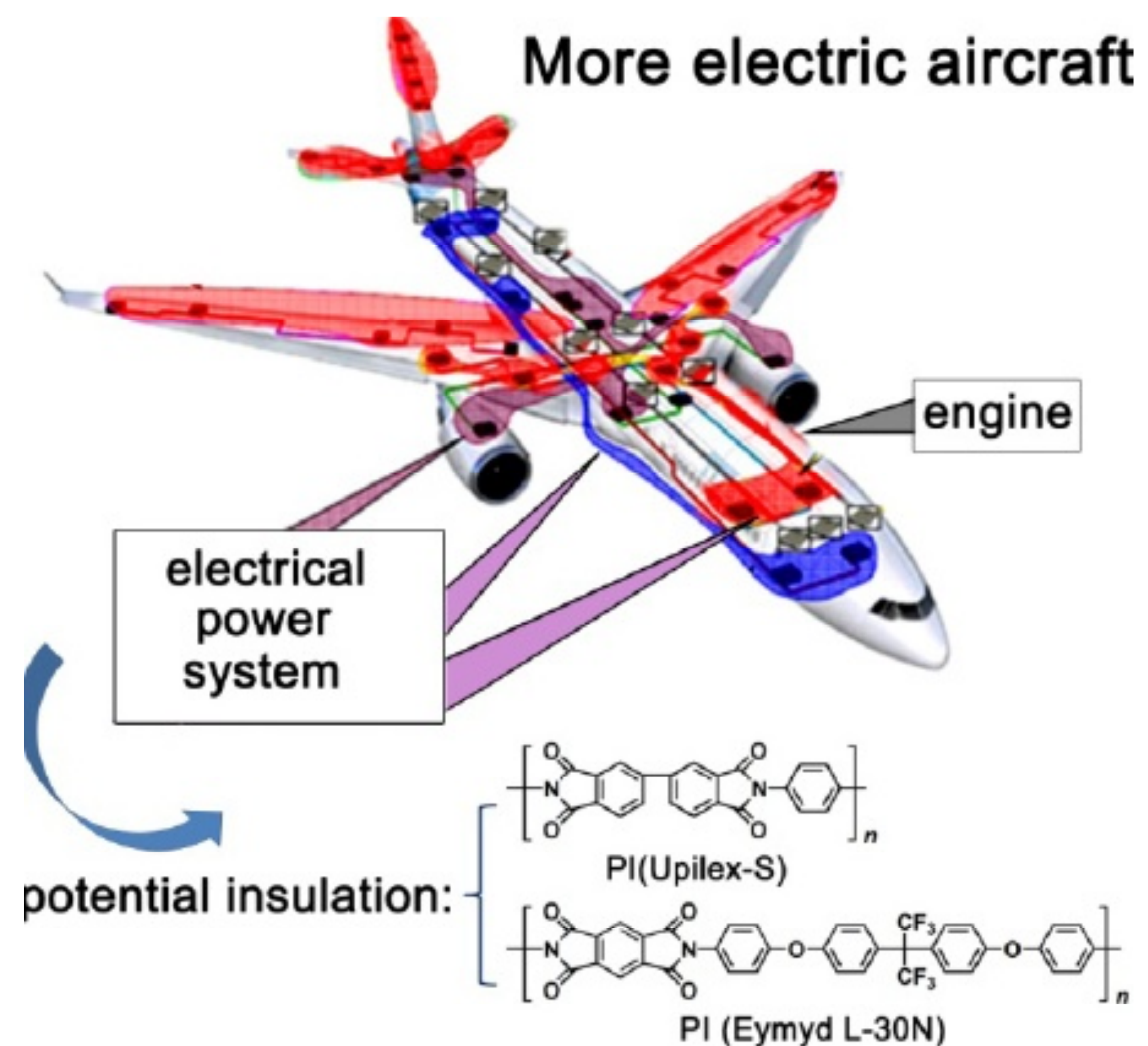

Fig. 6. Potential applications of PI dielectrics in MEA insulation.

Kapton® film. This makes it more useful for high voltage and high temperature aerospace wiring insulation applications. PPQ has also ever been evaluated as thermally resistant aerospace wire insulation in place of PI due to its superior toughness, thermal stability, low density (15% lower than that of PI), and especially hydrolytic stability [55].

For aviation application, recently, with the rapid development of aircrafts towards high-speed, lightweight, high reliability, and long servicing life, more severe properties are required to the electrical machines insulating materials. For example, the newly developed civil and military aviation motors have larger power requirements, higher power output, smaller volume, and lighter weight. Fig. 5 shows the development of power generation capacity for commercial aircrafts [56]. The increasing current density inevitably increases the working temperature for the electrical motors. Some aviation electrical machine can reach a long term servicing temperature of 250 °C, short term of 290 °C, and instantaneous temperature over 400 °C. Such a high-temperature environment requires the dielectrics possessing excellent thermal and dielectric stability.

Abdelhafez reviewed the development status of more electric aircrafts (MEAs) and their electric power generation systems [57,58]. MEAs (such as Airbus 380 and Boeing 787) achieve numerous advantages over conventional fuel-powered aircrafts, including improving the aircraft performance, decreasing operating and maintenance costs, and reducing the air pollutant gases [59]. However, MEAs put great pressure on the electrical system either in the amount of the required power and the processing and management of the power. Electrical insulation is a big concern due to the high power generation level. To make MEA the preferred option, the development of new dielectrics for components is necessary. For insulation materials, higher operating temperatures (up to or above 300 °C) are usually required. Upilex®-S PI film and Eymyd® PI film (a fluorinated PI film) are thought to be promising insulating dielectrics for MEAs electrical insulation system due to their good thermal stability and good resistance to humidity, ultraviolet radiation, basic solution and solvent at high altitudes [60,61], as illustrated in Fig. 6.

3.1.3. High-speed train electrical machines insulation

Wide application of adjustable-frequency and variable-speed techniques in high-speed train greatly promote the development of advanced HTPDs due to the specific property demands to the dielectrics for the application [62]. Generally, for the dielectrics using in high-speed train electrical machines insulation, excellent corona resistance, together with stable dielectric properties at high operating temperatures are highly desired. Conventional pure polymer dielectrics, such as Kapton® HN used for ordinary train electrical motor insulation usually cannot endure the corona damage and might lose the insulation ability under long-term electrical stress. In contrast, PI films with corona-resistant characteristics, such as Kapton® FCRC has been adopted in the European rail industry to improve the efficiency and durability of AC traction motors on high speed locomotives (such as Eurostar, etc.), where it significantly outperformed traditional insulation systems.

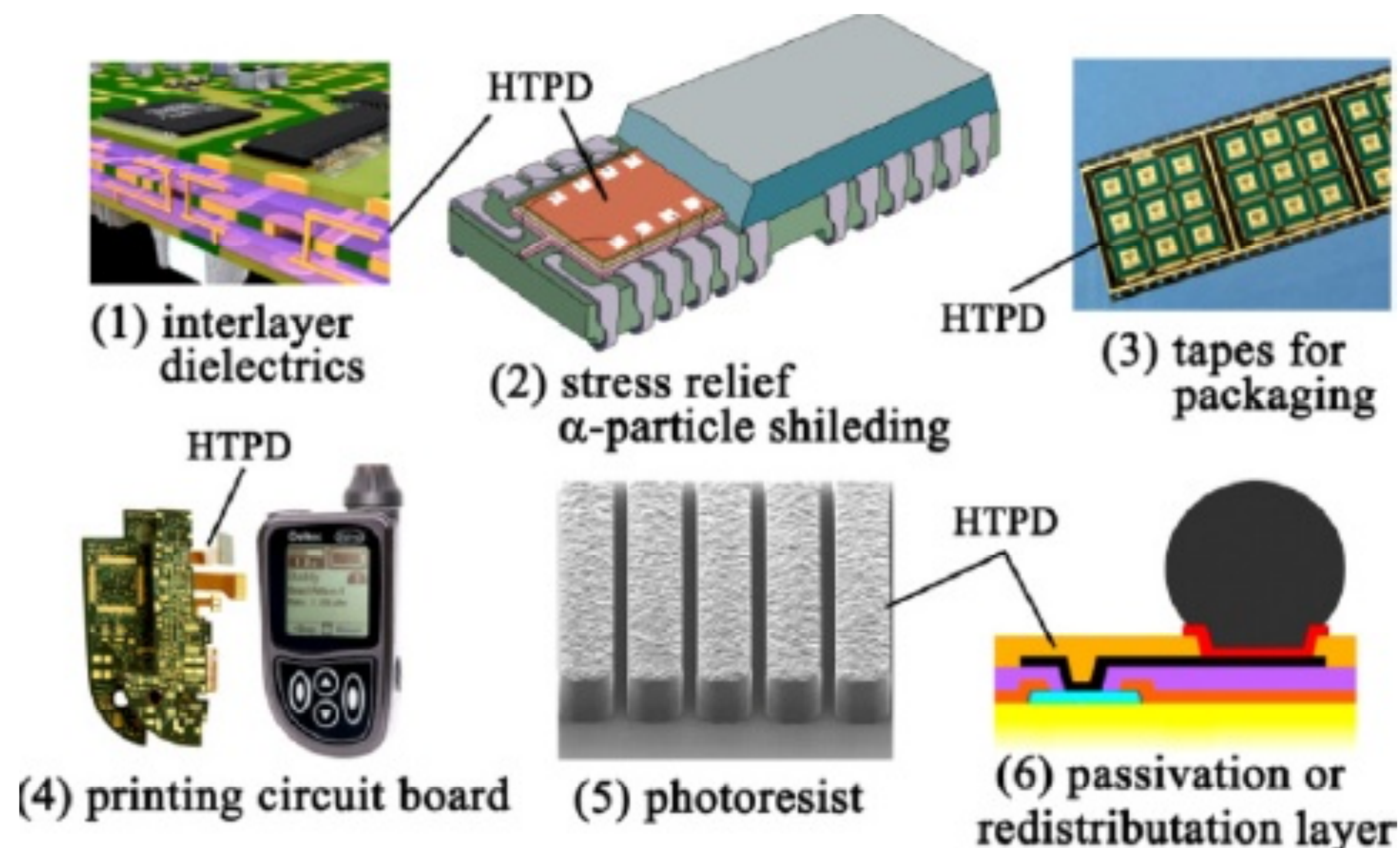

Fig. 7. Applications of HTPDs in microelectronic fabrications.

China is upgrading conventional railway lines and constructing tens of thousands of kilometers of high-speed railway lines. The total length of high-speed passenger lines will surpass 18,000 km within the next ten years, almost half of which will be 350 km/h lines. In the electrical machines, such as traction transformers for China Railway High-speed (CRH) electric multiple unit (EMU), common PI tapes, films and corona-resistant PI films (Kapton® FCRC) have been adopted as insulating materials [63]. In this area, numerous researches have been performed. For instance, Yang et al. investigated the corona aging in Kapton® HN PI film under alternative circuit (AC) voltage by Fourier transform infrared spectroscopy (FTIR) and the dielectric measurement [64]. It was found that the ether linkage, some of C-H bonds in aromatic rings and C-N-C bonds in imide were firstly broken by corona discharge. Then, with the increase of aging time, the cleavage of the imide group including C-N-C and C=O bonds occurs. At the same time, the oxidative degradation produced compounds such as carboxylic acid, ketones, and aldehydes. It has been well established that introduction of specific nanoparticles (alumina, titania, etc.) into the dielectrics can efficiently improved the corona resistance of the obtained nanodielectrics. This improvement is mainly attributed to the elimination of partial discharge, space charge, ionization, and other dielectric damage caused by the corona phenomenon. Zhou and coworkers studied and compared the charge transport mechanism in standard Kapton® HN film and corona-resistant Kapton® CRC film at a thickness of 0.025 mm [65,66]. The experiments indicated that the dielectric constant of Kapton® CR film is larger than that of Kapton HN, which enhances the barrier height for injecting charge into the dielectric, thereby increasing space charge accumulation threshold field. In addition, introduction of nano-particles raises the number of traps in Kapton® CR dielectric, thereby forming stable space charge electric field, which effectively increases dielectric properties of the film. Akram and coworkers prepared the PI/Al_2O_3 nanocomposite film by in-situ polymerization technique, together with the physical mixing (stir and ultrasonic) of nano-Al_2O_3 particle, followed by high-temperature imidization [67]. The degradation mechanism of nano-Al_2O_3 filled PI film due to surface discharge under square impulse voltage was then investigated. The results showed that failure of specimens is mainly caused by surface discharge which could be suppressed by the addition of nano particles. The nano-Al_2O_3 fillers improved the corona resistant property of PI film significantly. The corona resistant lifetime of PI/Al_2O_3 sample is six times longer than that of pure PI. Liu, et al. studied the effects of nano-Al_2O_3 contents on the dielectric properties of the PI/Al_2O_3 composite films [68]. They prepared the nanocomposite film by sol-gel method with aluminum isopropoxide as the precursor of nano-alumina in order to achieve a homogeneous dispersion of the nano particle. Experimental results revealed that addition of Al_2O_3 nanoparticles greatly improved the corona resistance of PI composite films. When the Al_2O_3 content was 12 wt.%, the corona resistance time reached 136 hours, while the pure PI film was only 0.5 hour. This improvement in corona resistance of PI/Al_2O_3 composite films should be attributed to the preventing charge accumulation effect of Al_2O_3 nanoparticles. Fan, et

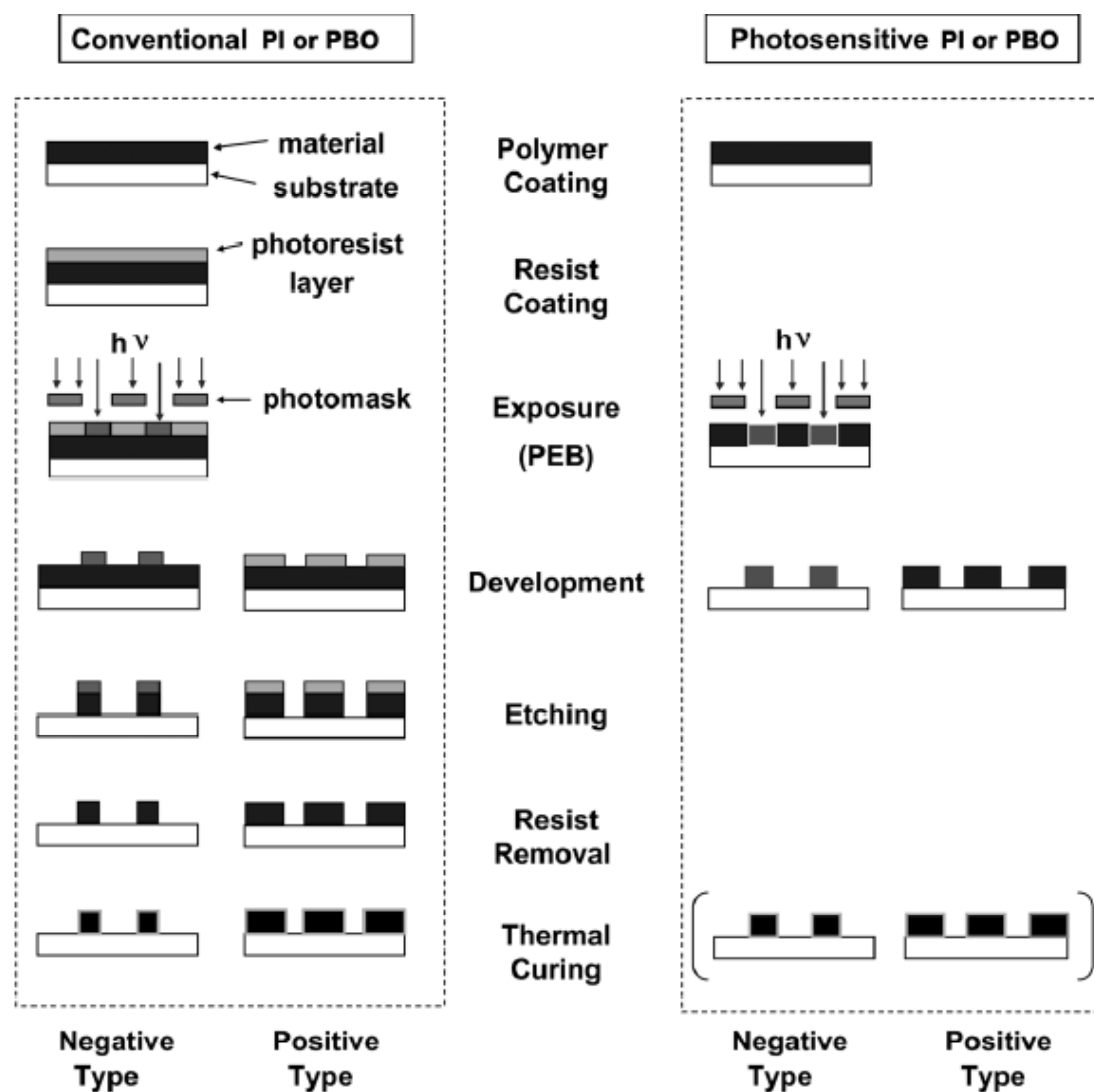

Fig. 8. Simplification of microelectronic patterning with photosensitive HTPDs. Reprinted with permission from T. Higashihara, Y. Saito, K. Mizoguchi and M. Ueda // *Reactive Functional Polymers* **73** (2013) 303, © 2013 Elsevier.

al. studied the impact of air relative humidity on the corona-resistant property of Kapton® CRC film [69]. It was found that the corona resistant aging for the PI film decreased logarithmically with the increase of the relative humidity. This phenomenon is closely relative with the intrinsically hydrophilic nature of the PI film.

3.2. Electronic insulation

Electronic industry, especially microelectronic and optoelectronic industry sees a tremendous development in the past decades. HTPDs materials, especially PI and PBO have good heat resistance, mechanical properties and electrical insulation; thus have been widely used in microelectronic and optoelectronic industry [70,71]. The typical applications of HTPDs in microelectronics are illustrated in Fig. 7.

PI and PBO chemistries deliver unique properties as interlayer dielectrics, redistribution layers, alpha-particle shielding layers, solder bump stress buffers, and passivation layers for microelectronic applications, including integrated circuit packaging, multi-chip modulus (MCM), microelectromechainical systems (MEMS), monolithic microwave integrated circuits (MMIC), etc. The polymers provide unique insulating protection and other functions to the electronic devices. More importantly, both PI and PBO chemistries offer flexible formulations of positive or negative photosensitive (or photodefinable) versions, which greatly simplify the microelectronic patterning, through-hole and other fabrication procedures without the aid of conventional photoresists, as shown in Fig. 8 [72]. Thereby, photosensitive HTPDs can obviously minimize the required facilities and realize a highly efficient productivity and low cost for microelectronic manufacturing. HTPDs with photosensitivity usually are double-purpose polymer dielectrics and are used as permanent materials or as temporary processing aids.

PI was introduced into electronics in early 1980s due to its suitable combination of high thermal stability, good mechanical properties, and a comparatively low dielectric constant. Lately, the rapid development of microelectronic industry desires dielectrics with lower dielectric constant, low moisture sensitivity, higher thermal and thermal-oxidative sta-

bility so as to achieve much rapid signal transmission speed. Then, PBO dielectrics attract much more attentions in recent years. Ueda and coworkers summarized the R&D progress for photosensitive PI (PSPI) and photosensitive PBO (PSPBO), respectively [73-75]. From these reviews, we can clearly find the developing course of PI and PBO dielectrics in microelectronic applications.

Very recently, PSPBOs with low curing temperatures have been paid much attention as dielectrics for microelectronic integration due to their good compatibility with modern microelectronic fabricating process, good environmental compatibility using an aqueous alkaline solution as a developer, together with their intrinsic excellent electrical, thermal and mechanical properties. Conventional PBO dielectrics are usually formed through cyclization of soluble PBO precursors, namely poly(*o*-hydroxyamide)s (PHAs) by thermal treatment over 300 °C. This high-temperature process is hardly applicable to some high-temperature sensitive integrated circuit devices, such as MEMS. Recently, the unique technique for low-temperature curing of PHAs is successfully overcome by modifying the PBO molecular structure and using the cyclization promoter, which greatly expands the wide application of PSPBO dielectrics [76]. Roberts compared the applications of high-temperature and low-temperature curable PI and PBO dielectrics in wafer-level chip scale packaging (WLCSP) [77]. WLCSP is thought to be the key technique achieving smaller and reliable mobile devices, such as smartphone, camera, tablet, etc. [78]. PI and PBO materials are recognized as the ideal dielectrics of choice for WLCSP fabrication due to their excellent thermal and chemical stability, relatively low dielectric constant and stress levels, and excellent mechanical properties. In addition, the commercialization of photodefinable versions for PI and PBO has enabled WLCSP by reducing the number of steps and enhancing the reliability of the final devices. Several HTPDs commercially available from HD-Microsystems, Japan including high-temperature curable PI (HD-4100, cure condition: 350-390 °C/1h) and PBO (HD-8820, cure condition: 300-350 °C/1h); and low-temperature curable PBO (HD-8930 and HD-8940) were compared in the investigation. It was found that the low-temperature curable PBO dielectrics could provide comparable dielectric strength, adhesion strength, mechanical properties, moisture absorption, and volume resistivity with the high-temperature counterparts. However, the WLCSP wafer warpage due to the thermal stress caused by temperature cycling for the low-temperature PBOs were much lower than those of the high-temperature ones due to the relatively lower curing temperatures less than 250 °C. Very recently, Nishimura and coworkers reported the 200 °C curable positive-tone photodefinable PBO (HD-8940, cure condition: 200 °C/1h) and evaluated its application for fan-out wafer level package (FOWLP) [79]. FOWLP technology is an enhancement of standard WLCSP developed to provide a solution for semiconductor devices requiring a higher degree of integration and a greater number of external contacts [80]. It provides a smaller package footprint with higher input/output (I/O) along with improved thermal and electrical performance. In FOWLP, it is preferable that the curing temperature of the buffer coatings and interlayer dielectrics is less than 300 °C for reducing the thermal damage of semiconductor devices and materials. In the evaluation, the PBO was cured at 200 °C for 1 h in nitrogen atmosphere. The experimental results indicated that the cure PSPBO exhibited excellent reliability against various reliability tests, including temperature cycle test (-55-125 °C, 500 cycles), temperature humidity test (85 °C/85% RH for 168 h) followed by 3 times reflow test (260 °C/10s). Thus, it might find various applications in FOWLP.

Photosensitive PI with ceramic nanoparticles fillers have also been widely studied for microelectronic applications [81]. The addition of nanosized ceramic fillers to a photoresist is driven by the following aspired aspects: improved sensitivity to electromagnetic radiation of a certain wavelength region; improved resolution; improved chemical resist stability; improved mechanical stability during processing; tailoring of the coefficient of thermal expansion; and introduction of new functionalities like electrical conductivity or magnetic properties. For example, surface modified nanosized SiO_2 (10–50 nm particle size) has been reported to be beneficial to improving the process stability of a photosensitive fluorinated polyimide due to the enhanced interaction of the particle large surface area and the polymer matrix [82].

4. CONCLUSIONS AND FUTURE PROSPECTS

As a class of high performance polymer dielectrics, HTPDs have been increasingly used in advanced electrical and electronic applications although they usually have much higher price than those of common polymer dielectrics. Thus, up to now, HTPDs are mainly used in special applications where their high cost is compensated by their special charac-

teristics. Nevertheless, HTPDs are attracting more attention and becoming indispensable for high-performance insulation. The foreseeable R&D trends for future HTPDs might include the following aspects. (1) Theoretically design and numerical simulation for HTPDs. Molecular design and property prediction via computational methods is undoubtedly attractive before large-scale production for HTPDs due to their high cost. Computational strategies for HTPDs design should be further explored and established [83].
(2) Low-cost preparation and manufacture procedures for HTPDs. Cost-effective synthesis and manufacturing procedures for HTPDs is of great concern for their wide applications in advanced insulation.
(3) Property modifications, functionalization and new materials R&D. Modification of the performance defects for the current used HTPDs is undoubtedly beneficial for expanding their applications. The topics include decreasing curing temperatures, improving moisture resistance, improving environmental compatibility, and so on. In addition, HTPDs with special function, such as super-lightweight, flexible and foldable, etc. are always highly desired in advanced insulation applications. For example, very recently, flexible and bendable high-temperature dielectric materials which can work stable at 250 °C have been reported [84].

In summary, HTPDs science and technology is an interdiscipline of polymer chemistry, material, electrotechnics, electronics and other related subjects; thus their development is highly dependent on the cooperation of scientists and engineers from different areas. With the rapid development of advanced electrical and electronic industry, HTPDs will see a bright future.

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