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Review

Modern approaches to the processing of bismaleimide resins

Sergey E. Evsyukov*, Tim Pohlmann and Matthijs ter Wiel
Evonik Operations GmbH, Gutenbergstraße 2, 69221 Dossenheim, Germany.

ABSTRACT

Bismaleimides (BMIs) belong to the class of thermosetting resins, also called addition polyimides. Introduced in the late 1960s, they have found numerous applications as matrix resins for high-performance, heat-resistant composites, adhesives, and coatings. The chemical composition of formulated BMI resins varies widely in order to meet stringent requirements being imposed upon mechanical and electrical properties of advanced composites. However, modifications are also necessary to meet the processing characteristics of both established and emerging composite manufacturing processes, sometimes at the expense of the resin properties. In this short review, modern approaches to the formulation and processing of heat-resistant bismaleimide resins are briefly discussed.

KEYWORDS: bismaleimide matrix resins, high-temperature thermosets, heat-resistant composites.

ABBREVIATIONS

BMIs	:	Bismaleimide(s)
CTE	:	Coefficient of thermal expansion
DABA	:	*o*,*o*'-diallylbisphenol A
DOPO	:	9,10-Dihydro-9-oxa-10-phosphaphenanthrene-10-oxide
HP	:	High-performance
HPT	:	High-performance thermoplastic
MATMD	:	1,6-bismaleimido (trimethyl) hexane
MEMDAB	:	bis(3-methyl-5-ethyl-4-maleimidophenyl)methane
MDAB	:	4,4'-bismaleimido diphenylmethane
MXBI	:	*meta*-xylylenebismaleimide
PMR	:	Polymerization of monomeric reactants
TDAB	:	2,4-bismaleimidotoluene
T_g	:	Glass transition temperature
TM123	:	4,4'-Bis(*o*-propenylphenoxy) benzophenone
TM121	:	1,1'-(1,3-phenylenedioxy)bis(3-(2-(prop-2-enyl)phenoxy) propan-2-ol)

1. Introduction

Composite materials constitute an already well-established, but still rapidly growing, cross-disciplinary branch of modern science and technology combining polymer chemistry, material science, and mechanical engineering. According to a market report by Grand View Research, Inc. [1], the global market size of composites is anticipated to reach USD 130.83 billion by 2024, while expanding at a compound annual growth rate (CAGR) of 7.8% over the forecast period. This growth is powered particularly by rising demand for lightweight, strong, and non-corroding materials from aerospace, automotive, wind energy, and oil & gas industries. The literature in the field of composite materials is abundant, and numerous excellent books have been published over the last decade [2-9].

Meanwhile, both thermosetting [8] and thermoplastic [9] resins have become established matrices for

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*Corresponding author: sergey.evsyukov@evonik.com

This review is faithfully dedicated to Dr. Horst D. Stenzenberger, the multifaceted scientist, passionate researcher, and creative inventor, whose pioneering studies paved the way for the development, establishment, and many practical applications of thermosetting bismaleimide resins.

composite materials. The global thermoplastic resin market in the composite industry is projected to reach USD 15.2 billion in 2024 [10]. Rapidly growing acceptance and use of composites in the automotive and aviation areas, rising demand for recyclable and fast-curing materials, coupled with increasing demand for lightweight components to address the pressing fuel saving standards are driving forces that make thermoplastic resins the fastest growing segment [1, 10, 11].

2. Thermosetting resins for advanced composites

Although thermoplastics offer high fracture toughness, composite damage tolerance, and recycling opportunities, it is the thermosets which still dominate the market for advanced composites [1]. Thermosetting resins were estimated to constitute the largest market for composites globally, with a share of 83.3% in 2015 [1] due to their easy processing, excellent mechanical strength, and heat resistance, ensuring, in contrast to thermoplastics, high shape stability at elevated temperatures, the properties being crucial in the aerospace area. The global market for thermoset composites is expected to rise to an estimated value of USD 80.81 billion by 2026, anticipating a CAGR of 6.77% in the forecast period [12]. The main issues restraining growth of this market are time- and energy-consuming cure and post-cure steps as well as limited or in many cases even zero recyclability and reusability of the cured products because of their three-dimensionally cross-linked structure.

The main types of thermosetting matrix resins being used in the production of advanced high-performance composites are listed in Table 1.

By volume, unsaturated polyesters are still the most dominant matrix resins for the production of glass fibre-reinforced composites. They are popular because of low price, ease of use, and reasonable performance characteristics; however, their temperature performance is rather moderate. Viscosities can be adjusted to meet composite processing techniques such as hand lay-up, wet-filament winding, pultrusion, and resin transfer molding.

For high-performance composites being increasingly used for advanced engineering and aerospace applications, carbon fiber-reinforced epoxy resins dominate the sector. Liquid bisphenol-A diglycidylether cured with diaminodiphenylsulfone provides an ultimate glass transition temperature (T_g) of 175 °C. Higher T_g's are achieved with polyfunctional epoxies such as tetraglycidyl methylenedianiline, triglycidylaminophenol, and glycidylated novolacs besides others, with a variety of curing agents such as polyphenols, polyamines, and anhydrides. Established processing techniques for carbon fiber composites were developed based on advantageous rheological properties provided by the epoxy resin formulations. Hot melt resin filming, followed by the deposition of collimated fibers or fabric onto the resin film to impregnate the fibers with the resin, and finally calendering of the lay-up between release papers to a uniform thickness is a widely used technique to manufacture prepregs (melt-prepregging).

As the development and the range of applications of advanced composites progressed, resins with higher use temperatures became a need. A variety of high-temperature thermoset resin chemistries such as multy-functional high-performance epoxy resins [23-26], cyanate esters [27-29], bismaleimides [30-32], benzocyclobutenes [33], benzoxazines [34-36], ethynyl-terminated oligomers [37-40], PMR polyimides (PMR = Polymerisation of Monomeric Reactants [41-45]), and phthalonitriles [46, 47] were developed to meet these requirements.

All these resins designed to withstand extreme heat and temperature environments, in particular under conditions of high humidity, constitute a branch of high-temperature matrix resins for the use in advanced composite applications. High-temperature resins have a long history in the composites industry, increasingly finding applications in the aerospace area, for both commercial and military aircraft engines [48, 49].

Growing acceptance of composites in the aerospace and automotive industries, coupled with a greater demand for light-weight composites, increasing production rates of high-temperature composite-rich aircraft, and superior performance benefits of high-temperature resins, are the key factors proliferating the growth of the market. Meanwhile, the global high-temperature composite resins market is projected to grow at a healthy rate over the next five years to reach USD 187.0 million in 2025, as estimated by Stratview Research [50, 51].

Table 1. Main types of thermosets used for advanced composites.

Resin type	Curing agent/ co-monomer/catalyst	T_g (°C)	Selected features	Refs.
Unsaturated polyesters	Styrene, acrylates, peroxides	60-140	Good fiber wetting, UV stability, low cost	[13-16]
Phenolic resins	Acids, metal salts	120-160	High char yield, low cost	[17]
Acrylate esters (also called vinyl esters)	Styrene, peroxides	120-180	Outdoor stability, corrosion resistance	[16, 18]
Vinyl ethers	Cationic initiators	80-145	Low cure shrinkage	[19, 20]
Epoxy resins, bisphenol-type	Aliphatic amines Aromatic amines Anhydrides	80-140 120-180 100-150	Good adhesion to fiber, low cure shrinkage, easy processing	[21, 22]
Multyfunctional HP-epoxy resins	Polyamines Novolacs, polyphenols	up to 250 180-230	Good hot/wet resistance, low toughness	[23-26]
Cyanate esters	Transition metal catalysts	180-300	Low dielectric constant, low CTE	[27-29]
Bismaleimides (addition polyimides)	Methylenedianiline, diallylbisphenols, bis(propenylphenoxy) compounds	250-270 250-320 250-320	Good toughness, high thermal and oxidative stability, hot/wet resistance, low CTE, low flammability	[30-32]
Benzocyclobutenes	Thermal cure	up to 300	Low dielectric constant	[33]
Benzoxazines	Thermal or acid-catalyzed cure	160-280	Low cure shrinkage, low water absorption, good flame retardance	[34-36]
Ethynyl and phenylethynyl-terminated oligomers	Thermal cure	240-360	Good thermal and oxidative stability	[37-40]
PMR polyimides	Thermal imidization and cure	340-400 and higher	High heat resistance and thermal and oxidative stability	[41-45]
Phthalonitriles	Thermal or amine-promoted cure; phenols, acids and metal complexes can also be used	350-420 and higher	High thermal stability and flame resistance; high post-cure temperature	[46, 47]

3. Bismaleimide resins

Among high-temperature matrix resins, bismaleimides (BMI) occupy a special position. Introduced in the late 1960s [52, 53], they quickly gained popularity and became very successful because their rheological properties could be adjusted to process them in an “epoxy-like” way, using essentially the same equipment [30-32]. Further benefits attracting end users include a balanced set of thermal and mechanical properties of cured BMIs, as well as their reasonable price-performance ratio. Numerous high-temperature applications, mainly in the aerospace area, have been developed since then, including engine nacelle components [49, 54, 55], bypass ducts [56, 57], thrust reversers [49, 58-60], and structural airframe parts of advanced combat aircraft and spacecraft [49, 58, 61-64].

A special and fast-growing area of composite tooling is also increasingly accommodating BMI matrices due to their low coefficient of thermal expansion (CTE) [65-71].

Formulated BMI resins are typically comprised of bismaleimide building blocks, co-monomers, and functional additives such as reactive diluents, tackifiers, thickeners, and tougheners.

3.1. Bismaleimide building blocks

Bismaleimide building blocks (BMI monomers) are organic compounds bearing two reactive maleimide groups of general formula shown in Fig. 1. In the case of BMI oligomers, their functionality can be >2.

Chemical structure of the spacer *X* may be varied over a wide range, and numerous BMIs have been reported so far in the literature [31, 32, 72, 73]. Typical BMI building blocks, currently adopted by the industry of composite materials, include 4,4'-bismaleimidodiphenylmethane (MDAB [74], Fig. 2,***1***), 2,4-bismaleimidotoluene (TDAB [75], Fig. 2,***2***), bis(3-methyl-5-ethyl-4-maleimidophenyl) methane (Fig. 2,***3***), 2,2-bis(4-maleimido phenoxyphenyl)propane (Fig. 2,***4***), and 1,6-bismaleimido(trimethyl)hexane (MATMD), the latter being an approx. 50:50 mixture of two isomers, namely 1,6-bismaleimido(2,2,4-trimethyl)hexane (Fig. 2,***5***) and 1,6-bismaleimido(2,2,4-trimethyl) hexane (Fig. 2,***6***).

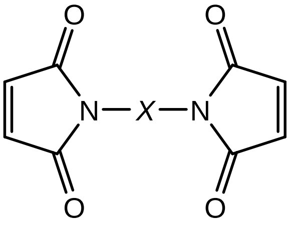

Fig. 1. General structure of a BMI monomer, where *X* typically represents an aliphatic, aromatic, or mixed aliphatic/aromatic spacer.

Despite the large number of new monomers synthesized over years that are claimed to offer improved processability or superior thermal and mechanical properties, the list of commercially available BMIs established in the industry is not that long due to pressing price-performance considerations. Finally, the toxicity of aromatic amines used as raw materials is becoming an increasingly important issue.

Although a variety of new formulated resins is regularly coming from laboratories, being reported in numerous patents and research papers, no new building blocks were commercialized for quite a long time. Actually, the only new BMI monomer that emerged in the marketplace within the last decade was *m*-xylylenebismaleimide (Fig. 2,***7***) recently launched as Compimide$^{®}$ MXBI [76]. Based on relatively inexpensive and low-toxicity diamine, this monomer shows lower melting point and better solubility as compared to ‘classical’ aromatic BMIs [77]. Formulated BMI resins based on MXBI were reported to show noticeably lower softening and melting temperatures as well as lower viscosity within the entire processing window as compared, for instance, with MDAB- and TDAB-based analogues [78-80] (see also Section 3.2. below).

3.2. Eutectics BMI mixtures

Aromatic BMIs like MDAB, TDAB, and MEMDAB are crystalline compounds with melting points as high as 156-164 °C, 172-178 °C, and 160-165 °C, respectively. Therefore, the melt processing of

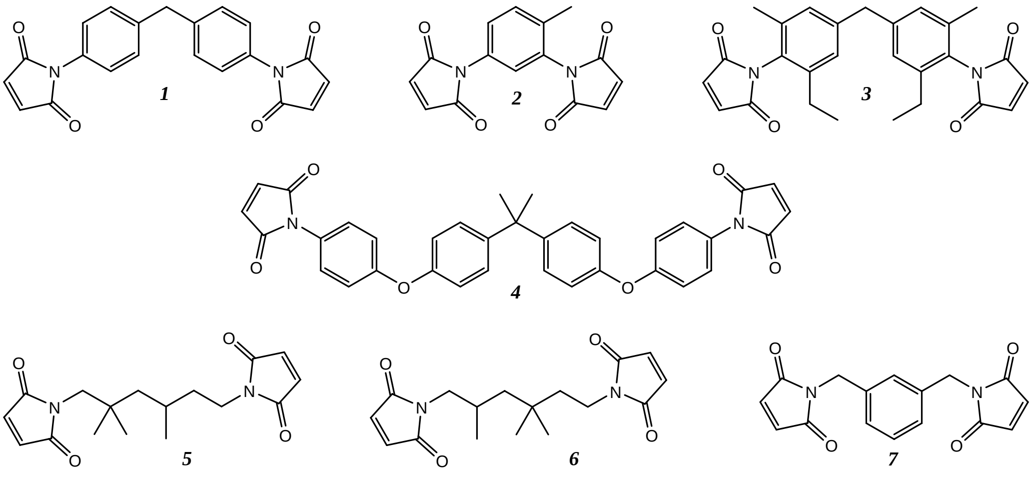

Fig. 2. Building blocks being used as a monomer in formulated BMI resins.

pure monomers requires high temperature, thus making this technology energy consuming.

In the early 1970s, it was found that binary eutectic mixtures of aromatic bismaleimides consisting of MDAB and TDAB blended with MATMD (Fig. 2,***5*** and ***6***) provided amorphous ternary eutectics with extremely low softening (70-90 °C) and melting temperatures (90-110 °C) and very low viscosities at temperatures above 110 °C [81, 82]. One of these resins was originally commercialized by Rhône-Poulenc in mid 1970s under the trademark Kerimide 353. Its advanced version Compimide® 353A is still available from Evonik [83]. It took another 15 years before researchers started investigating eutectic bismaleimide mixtures in order to better understand the basics of eutectic blends such as their melting characteristics, reactivity, rheology, and properties of their cured mixtures [73, 84-87]. Based on MXBI, a new low-melting, low-viscosity eutectic BMI resin blend Compimide® 50LM softening at 40-50 °C and melting at 80-100 °C was developed recently [79, 88].

3.3. Co-monomers

BMI monomers or their eutectic blends, when thermally cured in pure form, provide highly cross-linked, heat-resistant materials with T_g in excess of 320 °C. However, these materials are very brittle, and hence they cannot be used as matrix resins for structural applications in HP composites. Back in mid 1970s, it was found that blending of MDAB with *o*,*o*'-diallylbisphenol A (DABA) affords melt-processible resins, which after cure provide strong, tough, and still heat-resistant materials showing T_g in excess of 280 °C [89].

Since then, numerous allyl- (i.e. 2-propen-1-yl-) [90-105] and 1-propen-1yl-functionalized co-monomers [92, 97-99, 102, 105-107] have been synthesized. The reactive functionality mainly is based on 2-allyl- or 2-propenylphenol, eugenol or *iso*-eugenol as well as vanitrope derivatives [107]. Although co-monomers typically are difunctional compounds of the general formula ***I*** or ***II*** (Fig. 3), tris- and hexakis-derivatives based on tribenzoylbenzene [108-110], *s*-triazine [111, 112], and cyclotriphosphazene as core spacers [112-115] as well as polyfunctional oligomers like phenolic novolak [116], polysulfone [117], and poly (ether ether ketone) [118] have also been reported.

Typically, the reactivity of propenyl-functionalized co-monomers is higher than that of allyl derivatives. Therefore, cross-linking of BMI resins formulated with propenyl co-monomers begins 20-30 °C below the cross-linking temperature of formulations based on the allyl-type co-monomers, and the reaction occurs at a faster rate. On the other hand, the nature of spacers *Z* also plays a certain role. Generally, the electron-rich spacers like alkylated or alkoxylated phenylens or phenylene ethers provide higher reactivity as compared to electron-poor ones like benzophenone or diphenylsulphone, leading to faster curing systems. Recently, two new families of reactive co-monomers comprising alkenylphenoxy diphenylethylenes (Fig. 4,***a***) [119] and bisalkenyl (diphenyl ether)s (Fig. 4,***b***) [120] were reported to show higher reactivity as compared to commercial co-monomers.

On account of the cost considerations mentioned above, the list of commercially available co-monomers also is short, including two commonly used and well-established in the industry compounds, namely DABA [121] and 4,4'-bis(*o*-propenylphenoxy)benzophenone [122].

3.4. Reactive diluents

It is not an easy matter to draw a clear-cut line between co-monomers and reactive diluents. While co-monomers were designed for the use in BMI formulations, reactive diluents normally are common monomers also being used in other thermosetting

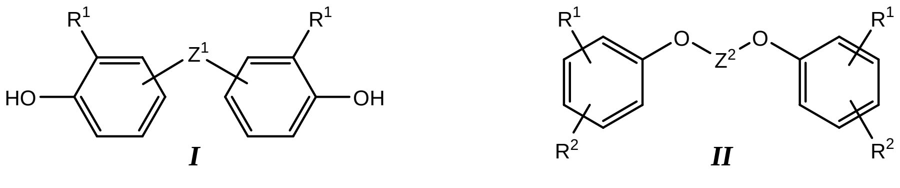

Fig. 3. General structures of difunctional co-monomers used in formulated BMI resins, wherein Z^1 and Z^2 are aliphatic, aromatic, heteroaromatic, or mixed spacers, R^1 is an allyl or propenyl group, and R^2 is H or an alkoxy group.

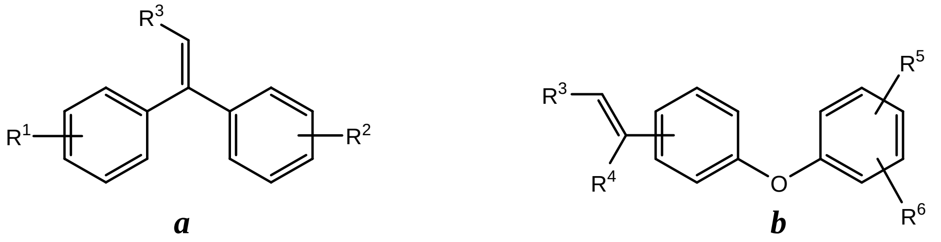

Fig. 4. Alkenylphenoxydiphenylethylene (***a***) and bisalkenyl(diphenyl ether) (***b***) families of reactive co-monomers, wherein R^1 is H or an alkenylphenoxy group, R^2 is an alkenylphenoxy group, R^3 and R^4 are H or alkyl groups, R^5 is an alkenyl group, and R^6 is H or an alkoxy group.

and thermoplastic polymers. Generally, reactive diluents are low-viscosity liquid compounds bearing unsaturated functional groups, capable of reacting with maleimide moieties. They are used in BMI formulations in order to reduce viscosity and T_g of uncured resins, thus facilitating the impregnation of fibers and improving drape and tack of prepregs.

Typically, allyl- and vinyl-functionalized compounds like allyl ethers [123-125] and esters [126-129], styrene and its derivatives [96, 130-134], divinylbenzene [124, 135-137], vinyl ethers [137-140], vinyl- and acrylamides like *N*-vinylpyrrolidone [128, 130, 137, 141, 142] and *N*-acryloylmorpholine [143] as well as acrylates [135, 142, 144, 145] are used as reactive diluents in BMI formulations. Polyfunctional compounds like triallyl cyanurate [146, 147] and isocyanurate [126, 127, 146-150] being commercially available [151] can also be used.

It should be noted that numerous liquid thermosetting monomers of a different nature like cyanate esters, epoxies, or diisocyanates that can be blended and co-cured with BMIs to form interpenetrating networks are not considered in this review.

The most frequently used reactive diluent for formulated BMI resins is diallyl ether of bisphenol A [78, 123, 125, 129, 130, 152-155], which is a commercially available raw material for industrial applications [156]. It should be noted that this compound is a precursor for the production of DABA co-monomer *via* thermal Claisen rearrangement [157]. Therefore, during cure process, diallyl ether partially transforms into DABA that can further react with maleimide moieties (see Section 3.8).

When choosing reactive diluents for BMI resins, one should also take into account kinetic and mechanistic aspects. For instance, vinyl and acrylic compounds are prone to radical homopolymerization, thus affecting the cure mechanism and driving T_g of the cured resin downwards.

3.5. Flow control agents

Some processing techniques like compression molding or open mold layup require adjusted resin flow during the cure step. If the viscosity of a binder is too low, the resin will just flow out of a laminate, thus deteriorating mechanical properties of a cured part. Therefore, a controlled advancement or thermal pre-polymerization of the binder called B-staging is normally applied to adjust rheological and also kinetic properties of the matrix resin.

Alternatively, or additionally, flow control agents can be used. One approach is using high-molecular weight thermoplastics like poly(ethylene oxide) [158, 159] soluble at elevated temperatures in the BMI resin melt, thus increasing its viscosity. Another approach is based on admixing fine particulate fillers. The latter can be either finely ground high-performance polymers like P84® polyimide [160] or inorganic particles like nano-silica [69-71, 161, 162].

A special type of particulate flow-control fillers are thixotropic thickeners that can be dispersed in the resin to adjust its rheology [163-165]. Thixotropic resins show low viscosity under the action of shear forces but become thick again at rest. Thus, Compimide® 50RTM, a formulated, low-viscosity BMI resin [80, 166] shows viscosities below 300 mPa·s at temperatures above 100 °C, and it already starts flowing away at temperatures as low as 50-60 °C. When modified with 6 wt.-% of thixotropic Aerosil® 200 [167], a piece of resin fully retained its shape after cure for 4 h at 170 °C.

3.6. Flame retardants

Although BMI resins show good fire-resistant properties by themselves [168, 169], various flame retardants are used to further improve them. Numerous inorganic fillers like silica or metal oxides, as well as halogen-, phosphorus-, nitrogen-, silicon-, boron-, antimony-, aluminum-, and magnesium-containing compounds were reported to be useful flame retardants for BMI resins [170]. In the past, halogen-containing compounds were widely used, but nowadays their application has been strongly reduced due to emission concerns, whereas phosphorus-containing compounds increasingly find application in BMI formulations. 9,10-Dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO) is a typical, commercially available flame retardant, being also used in BMI resins [171-173]. It should be noted that DOPO reacts with a double bond of the maleimide group *via* Michael addition (Fig. 5), thus covalently binding to the cross-linked polymer network in a cured composite [174-177].

In addition, phosphorus-containing co-monomers [112-115] and BMI building blocks [178-183] have also been reported to improve fire-resistant properties in formulated resins.

3.7. Tougheners

As mentioned above, pristine BMIs upon cure result in very hard, high-T_g, but brittle materials. Although the use of co-monomers has significantly improved the mechanical properties of cured BMI resins, the search for thermoplastic-like toughness, while retaining high T_g inherent in thermosets, has always been a kind of Holy Grail of composite engineers. However, when considered chemically, it should be noted that these two parameters in fact are antipodes, both being governed by the network density. Indeed, on the one hand, the higher is the cross-linking density the higher is T_g of a cured thermoset. On the other hand, the denser is the network, the lower is its flexibility and hence the toughness.

Increasing demand for high-performance, damage-tolerant composite structures driven primarily by the aircraft/aerospace industry, led to the development of toughened thermosets. Various techniques have been reported, while the approach of incorporating tough high-performance thermoplastics has become a common practice. However, the techniques to combine fibers with these high viscosity complex thermoset/thermoplastic systems (impregnation) had to be redesigned.

Generally, toughening additives form solid, elastic, or thermoplastic micro- or nano-domains evenly dispersed in discrete form throughout the matrix resin to increase its resistance to the crack-growth initiation. Various toughening mechanisms describing fracture behavior based on characteristics of viscoelasticity, shear yielding, and dilatational deformation involving cavitation were developed over the years [184, 185]. Basically, the toughening action consists in absorbing and dissipating energy concentrated at the forefront of a growing fracture, thus stopping its propagation.

Scientific literature dealing with this problem in thermosets is abundant [184-187], and numerous attempts have also been made to improve toughness of BMI resins [96, 185, 188-190]. Typical tougheners used in BMI formulations include inorganic nanoparticles like silica [70, 71, 155, 191-195] or silicon nitride [196], nano-clays [197, 198], as well as carbon [199, 200] and silica nanotubes [201], high-performance thermoplastics like poly(ether ketone)s [152, 153, 202-212], polysulfones and poly(ether sulfone)s [117, 154, 185, 209, 213-218], polyimides [84, 159, 218-223], poly(ether imide)s [185, 213, 214, 217, 224-229], polyesters [230-233],

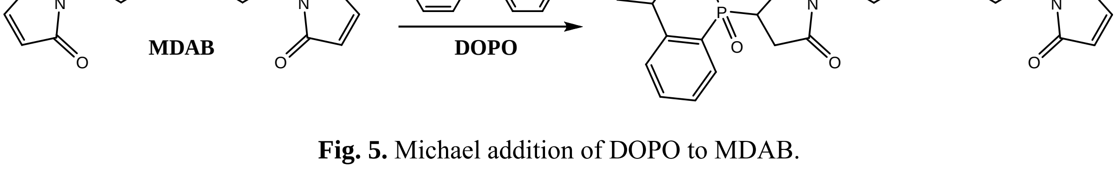

Fig. 5. Michael addition of DOPO to MDAB.

poly(ester imide)s [234], polyhydantoins [213, 214], and elastomers like acrylonitrile-butadiene rubber, carboxy- [235-237], vinyl- [238], acrylate- [239], or maleimide-terminated [226, 240]. Generally, the application of macromolecular tougheners involves their dissolution in a hot polymer melt followed by the phase separation during its cure with the formation of nano-droplets of the toughener, being in effect toughening elements, which are strongly affected by the concentration and molecular weight of the toughener. Reactive end-groups were reported to significantly contribute to the improvements in fracture toughness by covalently linking the discrete and continuous phases [218].

Another approach, that does not imply dissolution, employs micronized HP polymers like polyimide P84® [84, 158, 241], poly(phenylene oxide) [242], or polymeric core-shell nano-particles [241, 243].

Unfortunately, the use of all tougheners, excluding inorganic particles, leads to a decline in T_g, and all of them, to a greater or lesser extent, bring about increased viscosity of the resin melt, thus making their processing difficult. Therefore, the task of composite designers is finding a reasonable balance between coveted toughness and T_g, depending on the requirements of each particular application.

In order to overcome the viscosity issue, a special technique called interleaf or interlaminar toughening was developed [188, 190, 229, 244] in which thermoplastic layers are provided between the reinforcing layers at the layup stage. The thermoplastic layer that can be applied in a form of a porous film, fiber, or mesh is then dissolved during the cure step, finally providing an overall tough matrix.

Taking into account that toughness, being an ability of materials to resist fracturing under stress, is crucial for the impact strength of composite materials, whereas requirements imposed upon thermal and mechanical properties of composite parts are getting higher, the search for novel efficient toughening techniques remains a pressing endeavor.

3.8. Curing chemistry of BMI resins

Various chemical pathways are involved in thermal curing processes of BMI resins, depending on their composition. At high temperatures, typically above 180 °C, pure BMI building blocks undergo simple radical polymerization of their double bonds [245-248].

These radical reactions bring about a very high cross-link density and, as mentioned above, lead to the formation of very brittle materials, which do not find practical application.

In the case of the so-called chain-extended BMIs, being in fact Michael-type adducts [249, 250] of typically aromatic amines like methylenedianiline [96, 189, 215, 251-255], aminobenzhydrazide [256, 257], or bisthiols [258], the cure mechanism primarily is based on the Michael-type reaction of secondary amino-groups with terminal maleimide groups, although radical reactions also can be involved at the profound curing stages.

The mechanism commonly adopted for the cross-linking chemistry of formulated BMI resins [30-32] is based on a sequence of two chemical reactions, namely the *ene*-reaction, also referred to as the Alder-*ene* reaction [259, 260] and *[4+2]*-cycloaddition, broadly known as the Diels-Alder reaction or the Diene Synthesis [261, 262].

3.8.1. Allylphenol co-monomers

In case of the allylphenol-type co-monomers like DABA (Compimide® TM124 [121]) or Compimide® TM121 [98, 102, 263], the cross-linking sequence includes the Alder-*ene* reaction followed by the Diels-Alder addition [90, 264-274] (Fig. 6).

This cross-linking mechanism could be verified in model compound studies [266] with excess of the maleimide compound, but it was not found in the BMI/diallylbisphenol-A resins if their stoichiometry was close to 1:1 [267]. Based on IR, NMR, GCMS, and UV-reflection spectroscopic and calorimetric studies, the cure mechanism was found to include, apart from the Alder-*ene* sequence mentioned above, a complex combination of stepwise and chain polymerization, isomerization and re-arrangement/aromatization reactions, particularly at a later stage of curing, when mobility of functional groups is heavily restricted [267, 268, 270]. The opinions concerning these reactions are rather controversial. Rozenberg *et al.* [270, 271, 273, 275] suggested that cross-linking also can include dehydrative coupling of phenolic hydroxide groups. Some other groups argued in the same way [265]. However, allylphenol when heated alone did not form

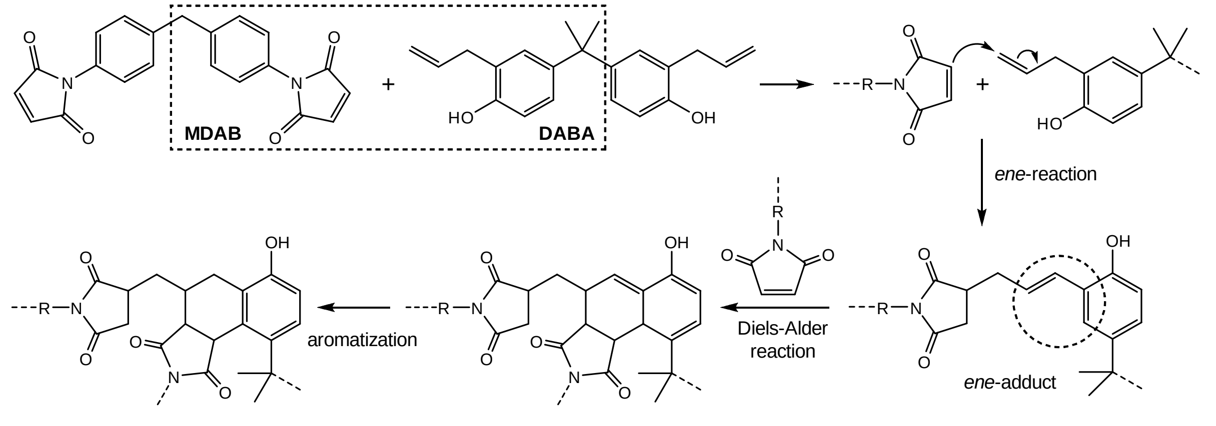

Fig. 6. Cure reactions of BMIs with allylphenols exemplified by the interaction of MDAB with DABA.

diallyldipenylether, and DABA did not homopolymerize *via* dehydrative polycondensation as well [265]. On the other hand, Michael addition of phenols to the electron-poor maleimide double bond, typically in the presence of bases, is well documented [276]. Finally, allyl groups can also contribute to the cross-linking during final curing steps [268].

3.8.2. Propenylphenoxy co-monomers

In case of the propenylphenoxy-type co-monomers like those shown in Fig. 7 [95, 98, 102, 107, 213, 215, 277], the cross-linking steps are considered to proceed the other way around. First, the Diels-Alder addition of the maleimide moiety to the propenylphenoxy group occurs, followed by the Alder-ene addition of the second maleimide group to the double bond formed in the first step (Fig. 8).

Indeed, early studies with *N*-phenylmaleimide and 4-(2-methoxy-4-propenylphenoxy)benzophenone [278] and with *N*-butylmaleimide and styrene [279] as monofunctional model compounds taken, correspondingly, at a 2:1 molar ratio did verify this mechanism. However, the mechanism has not yet been confirmed for resins based on difunctional bismaleimides and bispropenylphenoxy co-monomers. Possibly, sterical reasons may hinder or even prevent the postulated Alder-*ene* reaction in the second step of crosslinking shown in Fig. 8. Generally, the overall pathway of the reaction depends on the molar ratio of the components along with their structural features and thermodynamic parameters of the reaction. In the case of an excess of the BMI component, a Wagner-Jauregg reaction also can contribute to the cross-linking, which is a tandem Diels-Alder addition of two maleimide groups to one mole of the co-monomer like 1,1-diphenylethylene or styrene [280, 281]. Thus, the Wagner-Jauregg-type polyaddition of BMIs to 1,1-diphenylethylene taken in an equimolar ratio gave rise to the formation polyimides [282].

Although the curing of BMI resins on industrial scale is commonly performed by thermal cross-linking involving the mechanisms described above, alternative curing techniques including UV- [140, 141, 143, 283-289], electron beam- [290-297], and microwave-induced cross-linking [298-306] are also explored.

Generally, the UV-induced curing occurs at relatively low temperatures of 30-50 °C *via* radical mechanism [140, 143]. It was found that classical aromatic BMI building blocks like MDAB do not polymerize under UV light. In order to be cured they need a photoinitiator [140, 143]. In contrast, aliphatic BMIs were found to polymerize in the absence of photoinitiators due to the proposed abstraction of protons from aliphatic moieties under UV-irradiation [143, 286]. Because almost all BMI building blocks are crystalline or amorphous solids, the UV-curable formulations are normally based on liquid co-monomers like acrylates [287], acrylamides [143, 283], vinyl ethers [140, 286, 288, 289], styrene [288], or *N*-vinylpyrrolidone [288]. In case of aliphatic formulations, the so-called dimer BMI (also referred to as X-BMI, Q-Bond, or BMI-689), was also used, which is a viscous liquid at room temperature [284, 286, 289]. According to the proposed

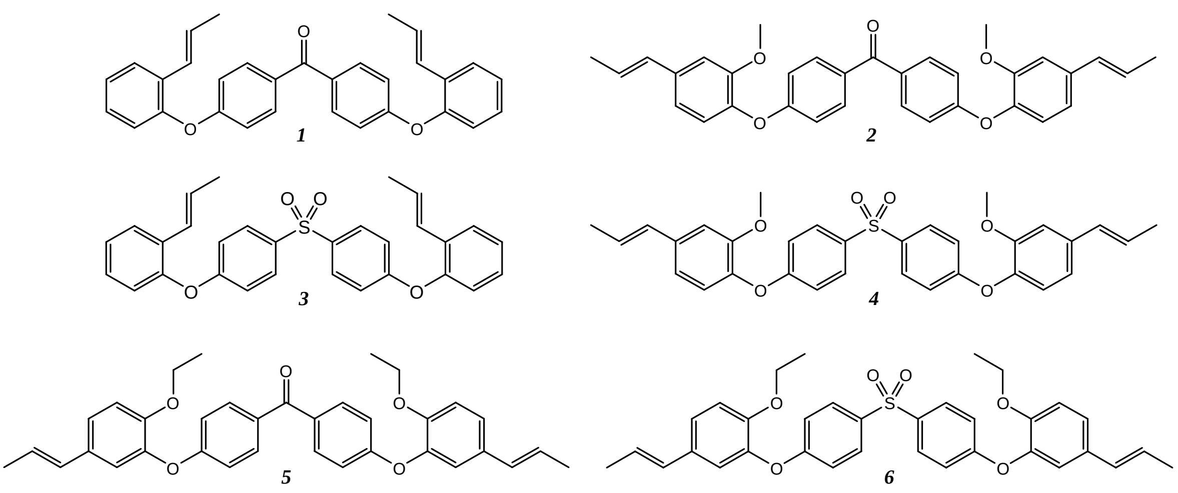

Fig. 7. Propenylphenoxy-type co-monomers being used in formulated BMI resins. ***1*** – 4,4'-bis(2-propenylphenoxy) benzophenone (Compimide® TM123 [122]), ***2*** – 4,4'-bis(2-methoxy-4-propenylphenoxy) benzophenone (Compimide® TM123-1), ***3*** – 4,4'-bis(2-propenylphenoxy)diphenylsulfone (Compimide® TM122), ***4*** – 4,4'-bis(2-methoxy-4-propenylphenoxy)diphenylsulfone (Compimide® TM122-1), ***5*** – 4,4'-bis(2-ethoxy-5-propenylphenoxy) benzophenone (Compimide® TM123-7), ***6*** – 4,4'-bis(2-ethoxy-5-propenylphenoxy) diphenylsulfone (Compimide® TM122-7).

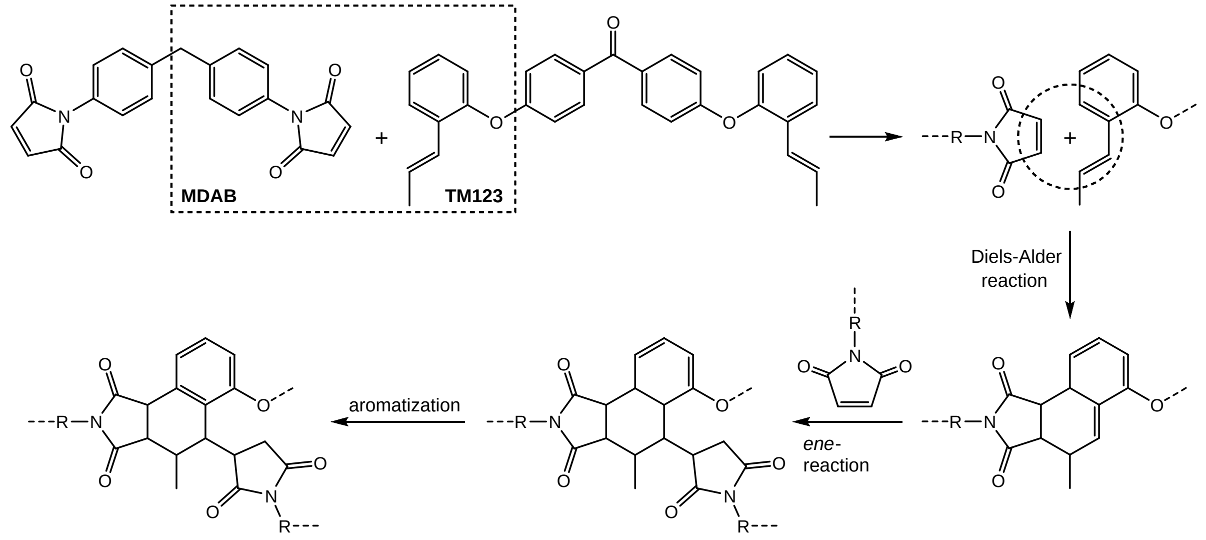

Fig. 8. Cure reactions of BMIs with propenylphenoxy-functionalized co-monomers exemplified by the interaction of MDAB with TM123.

mechanism [143], an alternating co-polymerization of BMI with a co-monomer occurs in the early stage of the curing to form linear and branched oligomers. As polymerization proceeds, the system gets cross-linked to form three-dimensional network, which can further be post-cured either photo-chemically or thermally.

Since penetrability of UV light into strongly absorbing organic media is limited to a few millimeters [307], the application areas of the UV curing typically include thin layer technologies like coatings and adhesives. Still, a multilayer approach being employed in the additive manufacturing can also be envisaged (see section 4.6).

In contrast to the UV irradiation, the electron beam-induced curing allows one to make reinforced composite materials up to 24 mm thick, depending on the beam energy, due to deeper penetration of electrons [308]. This thickness is limited by the maximum accelerator energy of approx. 10 MeV in order to avoid an induced radioactivity in the target materials. Nevertheless, thicker parts can be produced by stepwise, layer-by-layer curing approach that does not require high electron intensity [309].

Similarly to the UV-curable formulations, the electron beam-curable systems also comprise liquid reactive diluents like *N*-vinylpyrrolidone [292-297] or styrene [290, 295]. It was reported that classical BMI resins like systems based on MDAB, TDAB, and DABA do not polymerize at low electron beam intensities at temperatures below 100 °C [292-295]. Typically, no radical initiators are required, but the use of reactive diluents along with initiators allows one to cure BMI formulations at low intensities and low temperatures. The curing at higher beam intensity of 40 kGy per pass brought about the conversion degree above 75% with simultaneous heating of the matrix up to 250 °C due to combined action of the absorbed electron energy and the heat released by exothermic polymerization reaction [292-295], thus shifting the process towards conventional thermal curing. Generally, the advantages of the electron beam curing compared to thermal curing include lower thermal stresses in the cured resin, lower energy consumption, lower emission of volatiles, reduced curing time, and longer shelf life of the initiator-free formulations [308, 309].

Still another curing technique based on the microwave-induced cross-linking is a kind of hybrid technology, in which the polymerization reaction is induced thermally, whereas the heating is performed by the microwave irradiation [298-306]. The microwave heating allows one to quickly attain processing temperatures, being uniform throughout the entire bulk of the composite material, whereas nearly 95% reduction of cure-induced strains could be achieved as compared to the conventionally cured parts [303].

3.8.3. Curing catalysts

The curing process of formulated BMI resins can be accelerated with the use of various catalysts, which include peroxides, tertiary amines like triethylamine, dimethylbenzylamine, or 1,4-diazabicyclo[2.2.2]octane [256, 310], imidazole derivatives [256], phosphines and phosphonium salts [311, 312]. Peroxides promote radical cross-linking and allow reduction of the curing temperature. However, as mentioned above, radical cure results in the formation of brittle materials. Organic bases are known to catalyze Michael addition of primary and secondary amines and phenols to the maleimide group. Therefore, they are efficient promoters for the chain-extended BMI resins. Triphenylphsophine was shown to reduce gelation time of a BMI formulation based on MDAB and DABA by a factor of four, whereas tertiary amines did not show catalytic activity. It should be noted that a ^{13}C NMR study of model reactions involving monofunctional phenylmaleimide and 2-allylphenol has revealed that triphenylphosphine predominantly catalyzed trimerization of maleimide groups [312]. Unfortunately, no efficient catalysts have been found so far that would significantly accelerate the Alder-*ene* addition. Since catalysts speed up the cross-linking reaction, their type and concentration affect the cure time and the resin flow characteristics [310]. On the other hand, catalysts also can adversely affect processability of the BMI resins, reducing their shelf and pot life. Therefore, the type and concentration of the catalyst should be carefully tuned in order to reach a reasonable balance between the shelf life, pot life, and processing temperature.

4. Composite processing techniques

Technologies established for the manufacture of fiber-reinforced composites are manifold [3, 313-317]. What is described with the term “processing technique” is the sum of at least three individual process steps, i.e. i) impregnation of the reinforcement with the binder resin, ii) fiber or prepreg placement, and iii) cure of the matrix resin.

As it was mentioned above, formulated BMI resins combine low melt viscosity with a sufficient pot life, thus making them adaptable to most of the common processing technologies developed for the processing of conventional thermosetting binders, primarily epoxy resins. Typical technologies include hot melt prepregging [318-326] (whereas ready-to-use prepregs have become commercially available products [327-333]), compression molding [334-337], resin transfer molding (RTM) [338-344], various types of vacuum-assisted resin infusion [80, 345, 346], pultrusion [347-350], and filament winding [73, 81, 133, 263, 351-357].

In some techniques, such as wet filament winding and pultrusion, the impregnation of fibers with the thermoset resin is performed *in situ* just before shaping/molding/curing the final part. The sequence of the three process steps can vary, depending on the composite processing technique.

4.1. Resin transfer molding (RTM)

Generally, RTM is described as a technique in which the dry reinforcement, preferably as a preform, laid into a closed mold cavity, is impregnated with a low-viscosity resin that is subsequently cured. Within the vacuum-assisted processing technologies, the resin is transferred into the mold or infused into the pre-form by vacuum. In a variant of the RTM process called vacuum assisted resin infusion (VARI), one half of the mold is replaced by a flexible foil (vacuum bag) [80]. It should be noted that the VARI technique in various sources is also referred to as Vacuum Assisted Resin Infusion Molding (VARIM), Vacuum Assisted Resin Transfer Molding (VARTM), Vacuum Bag Resin Transfer Molding (VBRTM), Seemann Composites Resin Infusion Molding Process (SCRIMP), all of them being in effect the same technology, which implies infusion of liquid resin into a dry reinforcement by the vacuum [358, 359].

Historically, the RTM process became viable as a low-cost technique for the production of non-aerospace glass fiber-reinforced polyester composites in the early 1970s [360]. In the mid 1980s, improvements made in the injection equipment, mold design, and resin chemistries gave rise to the production of aerospace-quality components based on a range of thermosetting resins, including BMIs [342-344, 361, 362], and various reinforcements.

Studies dealing with RTM in general [339, 363, 364] and with RTM of bismaleimide resins [342-344, 361, 362] refer in particular to the requirement of a low resin viscosity for the process. Low viscosity is of main importance to guarantee proper wet-out of the fibers and sufficient pot life (2-4 hours), which is necessary when large parts are to be fabricated. Resin viscosities ranging from 150 to 1500 mPa·s are considered as most appropriate to manage short mold filling times and good fiber wet-out. Concerning the BMI resin formulations, low viscosities are rather difficult to achieve because of the inherent properties of raw materials making up the resin. Thus, the BMI building blocks typically are high-melting solids, and the co-monomers are semisolids or liquids with high viscosities at room temperature. Therefore, temperature normally is the tool to adjust the required process viscosity. Low toxicity certainly is among very important properties to provide a clean and safe production environment.

Presently, one-component, ready-to-use formulated BMI resins are commercially available for the RTM processing [166, 365].

4.2. Out-of-Autoclave (OoA) curing

Traditionally, the curing of thermosetting composite parts is performed in autoclaves, which provide both high temperature and uniform pressure of 3-14 bar (typically 4-7 bar), to compact (debulk) plies and fibers of the reinforcement and to suppress the formation of voids caused by the entrapped air and volatiles [366]. As composite parts, particularly for aerospace applications, are getting larger and larger, their autoclave cure becomes cumbersome. Modern autoclaves are huge and complex devices that are expensive both in procurement and in maintenance. As an alternative, the out-of-autoclave (also called vacuum-bag-only, VBO) technology was developed [367-369]. The OoA technology implies laying prepregs on a mold (tool), covering and sealing layup with a heat-resistant flexible foil (bag), applying vacuum to the bag to debulk the layup, and finally curing the part in a conventional oven. This technique is less capital-intensive and less expensive to operate than an autoclave, particularly when manufacturing very large parts. Indeed, the use of the OoA technique within the Ares V cargo launch vehicle program was reported to allow savings estimated $100 million needed for a 12 m by 24 m autoclave [367].

Unlike autoclave processing, the OoA-VBO technique affords significantly lower pressure, typically about 1 bar. Therefore, a VBO-compliant matrix resin should have low viscosity before curing and low proportion of volatiles in order to reduce content of voids. Since its emergence in early 1990s, the OoA technology has matured to become capable of manufacturing parts of the “autoclave quality”, reaching content of voids below 1%. In the meantime, BMI resins were successfully adopted for this technology [370-374], and special prepreg grades were developed [333, 375].

A special kind of the advanced OoA composite curing technology uses a heat transfer fluid to apply heat and modest pressure to the uncured BMI prepreg during processing [376].

4.3. Towpreg filament winding

Tow prepreg (towpreg) filament winding is a technology that combines advantages of low-cost manufacturing techniques with the use of high-performance and high-temperature matrix resins such as BMIs. The key feature of the towpreg filament winding process is the separation of the fiber impregnation step from the part winding operation. The towpreg fabrication operation is performed by specialists, who have the expertise and equipment to blend and handle resins safely and impregnate fiber tows to meet tight specifications [351]. Several methods may be employed to impregnate the fiber tow with a resin.

4.3.1. The solvent-based process

For impregnation, this process utilizes formulated resins dissolved in organic solvents. The solution concentration and viscosity are adjusted to allow a defined resin proportion, which is essential for a controlled and uniform resin content in the final composite part, and for the part quality. Within this process, the fiber tow is guided through a resin solution tank, and then through a drying zone (drying tower) to strip off the solvent and to advance the resin to a degree that a flexible but dry product is obtained. Finally, the non-sticky tow prepreg is wound onto a bobbin, which can be stored for an extended period of time before use in the towpreg winding process. The process is advantageous with respect to the fast and uniform tow impregnation due to low viscosity of the resin solution (typically, 100-250 mPa·s).

Bismaleimide resin solutions, such as Compimide® 1206 [377] and Compimide® 1224 [378] designed for the manufacture of glass fabric prepregs for printed circuit boards (PCB), can also be used in the described towpreg process. Soluble high-performance thermoplastics (~10-20%) may be added to the resin solutions for toughening.

4.3.2. The hot melt process

In this case, the binder system is applied onto the tow at a temperature, at which the resin is a low-viscosity liquid. Some of the high-performance resins like polyfunctional epoxies, cyanate esters, and bismaleimides have high melting transitions and, therefore, require elevated melt impregnation temperatures. In the melt process, the fiber tow is guided over a roll, onto which a resin film of a controlled thickness is applied and uniformly flattened using a system of rollers [379] or a doctor blade [380] for impregnation. The impregnated tow may subsequently be B-staged in a heating zone to yield a flexible but non-sticky towpreg.

The process is suitable for the state-of-the-art bismaleimide resins formulated as homogeneous blends of bismaleimide building blocks like MDAB [74] and MXBI [76] with DABA (Compimide® TM124 [121]), and/or bis(propenylphenoxy benzophenone) Compimide® TM123 [122]). These formulated BMI/co-monomer blends show viscosities in the range of 80-250 mPa·s at temperatures of 80-125 °C, at which the film transfer impregnation can be performed. It should be noted that resin blends formulated with low-viscosity reactive diluents can remain liquid at room temperature. Furthermore, it is standard practice that formulated BMI matrix systems are modified with high-performance thermoplastics like polysulfone, polyetherimide, or polyimide to improve the toughness of resins and the impact damage tolerance of cured composites made thereof. The thermoplastics can be dispersed in the BMI resin melt as fine powders of size 30-80 microns with almost no detrimental effect on the impregnation characteristics of the resin [381].

4.3.3. The powder process

In the powder process, the resin particles sized 80-150 microns are dispersed in air to form a fluidized bed, while the fiber tow is spread in an air stream. Electrostatic charging of the resin powder facilitates its adhesion to the grounded fiber tow. After passing the fluidized bed, the tow with adhering resin powder is then guided through a heating zone to melt the resin and impregnate the fibers. Finally, the impregnated tow is flattened in a calender to the desired uniform thickness. Originally developed for thermoplastic powders [382, 383], the process can also be used with thermosets. Bismaleimide resins that show uncured T_g's or melt transitions between 75 and 125 °C are well suited for the powder technology. Thus, a slightly pre-polymerized reactive BMI resin called Compimide® P500, which was prepared by blending MDAB and Compimide® TM123 in an extruder [384], was ground to form a non-sticky fine powder melting in the range of 100-110 °C and successfully tried in electrostatic fiber coating processes.

4.4. Powder processing

Generally, parameters of the composite manufacturing process dictate the resin processing properties such as viscosity, pot life, gel time, etc. Matrix resins can be tailored to meet the processing requirements, but often at the expense of attainable physical properties of the cured resin. The BMI resins, possessing outstanding properties, are in most cases solids at ambient and even at moderately elevated temperatures, owing to the optimized molecular weight of a pre-polymer and/or high melting temperature of individual building blocks. Therefore, modern techniques employ powdered BMI resins instead of solutions or hot melts. The use of aqueous powder dispersions for fiber/fabric impregnation has been successfully demonstrated both on laboratory and pilot-plant scale. Glass fabric laminates molded at pressures up to 10 bars show mechanical properties equivalent to or better than solution-based systems. In particular, the high-temperature physical properties are improved due to the absence of any plasticizing residual solvent. Another strong argument for prepregging and molding techniques employing aqueous slurries is the elimination of toxic solvents increasingly raising environmental and health safety concerns.

The powdered BMI binders that can be applied in this technology are based on pre-polymerized resin systems like Compimide® 200 or Compimide® P500 [384]. The powdered resins can be suspended in water or water/ethanol mixtures, and such suspensions may be used in a standard solvent/solution-based prepreg system operation with a glass or carbon fabric as reinforcements. The problems that have not fully been solved yet include the insufficient suspension stability due to the particle sedimentation, and the poor adhesion of the powder onto the reinforcement. An optimized resin particle size along with the use of efficient co-polymerizable emulsifiers in low concentrations may solve these problems in the future. The latter should be carefully studied in detail, as the stabilizers may have adverse effects on thermal and/or mechanical properties of the cured BMI resin.

4.5. Film stacking

An interesting, although not widely used technology, is the so-called film-stacking. The reinforcement (preferably as fabrics) and the resin film are stacked alternatingly, and the lay-up is molded afterwards in a heated press at high temperature. This technique initially was developed for thermoplastic resins and, therefore, high pressure and high temperature were required to achieve sufficient flow during molding [385, 386].

It is also possible to use thermoplastic/BMI blends in the form of films instead of thermoplastics. Such BMI/thermoplastic blend systems containing an excess of thermoplastic resin allow the manufacture of very tough laminates, whereas their processing temperature is significantly below that of neat thermoplastic resins, because the BMI part of the blend contributes to higher flowability and lower cure temperature [387].

One can realize the importance of the bismaleimide/thermoplastic blends, taking into account the toughening and processing problems. As already mentioned earlier, brittle BMI systems can be modified with high-performance thermoplastic (HPT) powders to improve the resin fracture toughness and composite damage tolerance. Usually 15-20% of HPT is used so that the modified thermoset resins retain their processing characteristics, mainly low viscosity. The toughness increase in the cured system is a result of the phase inverted or co-continuous morphology, depending on the HPT concentration in the blend. For a particular bismaleimide/polyetherimide system, the fracture toughness and morphology spectrum were reported [225], showing complete phase inversion for 20-30% polyetherimide with a local maximum at this HPT content. Another technique employs a low-melting, low-viscosity BMI resin as a plasticizer for HP, high-molecular-weight polymers (not necessarily thermoplastics) such as polyimide, polyamidoimide, and polyhydantoine. Blends of polyimide or polyhydantoin with up to 30-35% of bismaleimide (Compimide® 353) could be processed to films containing uncured BMI acting as a plasticizer at elevated temperatures. When heated to temperatures of 120-200 °C, the film softened and started to flow, if pressure was applied. The cure was performed in the temperature range from 200 to 275 °C. Owing to this behavior, it is possible to use such "hybride" films in the "classical" film-stacking process to mold laminates. The only difference to thermoplastic film stacking/molding is the time-consuming thermoset cure.

The bismaleimide resins suitable for such applications include systems like MDAB/DABA or Compimide® 96/Compimide® TM123 blends, because they are soluble in organic solvents such as *N*-methylpyrrolidone, *N*,*N*-dimethylacetamide, and methylene chloride to be processed into film in combination with the polyimide and/or polyhydantoin. In order to obtain strong, free-standing flexible films with a high BMI content, the molecular weight of the HPT polymer should be high enough.

There are some other applications possible for "hybride" BMI/HPT films such as high-temperature film adhesives and films for BMI composite interleave toughening [388].

4.6 Additive manufacturing

Additive manufacturing, commonly known as 3D printing, has emerged as a disruptive technology that is capable of fabricating three-dimensional objects with complex geometries by creating materials in a layer-by-layer way [389, 390]. Developing at a very fast pace, this technology holds potential for the dramatic change of human life in the future. The World Materials Forum named 3D printing of plastics and carbon fiber composites among top ten technologies that can reach industrial scale by 2030 and can decouple economic growth from the current use of natural resources while creating value for the industry all along the global supply chain [391].

Recently, BMIs were claimed to be a promising raw material for 3D printing [141, 392-398]. Several studies reported the use of the so-called X-BMIs as a bismaleimide component of the printing ink [392-395]. The X-BMIs, sometimes also referred to as Q-Bond, are the class of BMI building blocks and BMI-terminated, imide-extended oligomers based on dimer diamines. In turn, dimer diamines are branched aliphatic, either saturated or unsaturated, compounds made of C_{36}- dimer diacid, which is a dimerization product of unsaturated fatty oleic and linoleic acids [399]. Developed in early 1980s [400], X-BMIs were re-introduced and further developed some twenty years later [401]. It is noteworthy that X-BMIs with molecular weights below 1700 $g{\cdot}mol^{-1}$ are high-viscosity (1.5 Pa·s) liquids at room temperature [393]. Based on X-BMIs, stable ink formulations containing up to 80 wt% of BMI, about 3 wt% of photoinitiator, and an organic solvent were prepared and used in a PolyJet 3D printing process that employs UV-assisted curing [392-394]. Thermal post-cure at 275 °C was carried out in order to improve the mechanical properties of the printed objects. Despite liquid state of the monomers, their viscosity was still much too high for the PolyJet process. Therefore, the use of organic solvent as a diluent was necessary. In an effort to further reduce viscosity, the printing was performed at moderately elevated temperatures of 55-70 °C. Since the solvents used in the ink formulations were non-polymerizable, special care was taken to remove the solvent after each printing run [394]. For this purpose, IR irradiation was used in addition to the UV source. It should be noted, however, that very long chains between maleimide groups in X-BMIs result in a low network density, which in turn causes low T_g values in cured resins [394]. The addition of 5wt% of MDAB to the ink formulation allowed a modest increase in T_g from 71 up to 93 °C [394], which is still far below values known for BMI resins.

In contrast to the inks formulated with X-BMIs and inert solvents, the use of polymerizable reactive diluents like *N*-vinylpyrrolidone (NVP) [141, 396] and multifunctional (meth)acrylates [397, 398] along with "classical" BMI building blocks and co-monomers afforded printable inks that after cure resulted in high-T_g materials. For instance, when a slightly advanced mixture of MDAB with DABA, diluted with NVP-containing photoinitiator, was 3D-printed under UV-curing, and the printed object was post-cured for 8 h in the temperature range of 150-240 °C, the cured resin showed T_g of 210 °C [141]. Furthermore, 3D printing with inks based on MXBI or MDAB and multifunctional (meth)acrylates gave rise to materials that after post-cure showed T_g in excess of 300 °C. Moreover, the use of eutectic BMI mixture of MXBI, MDAB, and MATMD instead of pure BMI building blocks along with TM123 as a co-monomer allowed significant improvement of the resin processability [397, 398].

5. Conclusion

Being in industrial use for almost half a century, bismaleimide systems remain popular binders and still dominate the field of heat-resistant matrix resins. Despite the long history, their technological potential has not been exhausted. In early years, BMI resins suffered from the image of hard processability and poor toughness. However, within the last two decades, suppliers have developed grades, which can meet both the processability and the toughness standards of epoxies. On the other hand, fabricators came up with modified and new approaches to the processing of thermoset/thermoplastic blend resins needed for damage-tolerant composites. The towpreg filament winding, resin-suspension prepregging, film-stacking, and 3D printing increasingly find their way in the production. These techniques, being well suited for bismaleimide-based high-performance composites, hold promise for future applications. Finally, the ever-increasing use of automation, robotization, and digitalization in the production of composite materials has meanwhile been extended into the area of BMIs. In summary, it can be concluded that BMI resins will continue to be utilized across numerous high-tech applications for many years to come.

CONFLICT OF INTEREST STATEMENT

The authors declare no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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