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Polymer Matrix Wave-Transparent Composites

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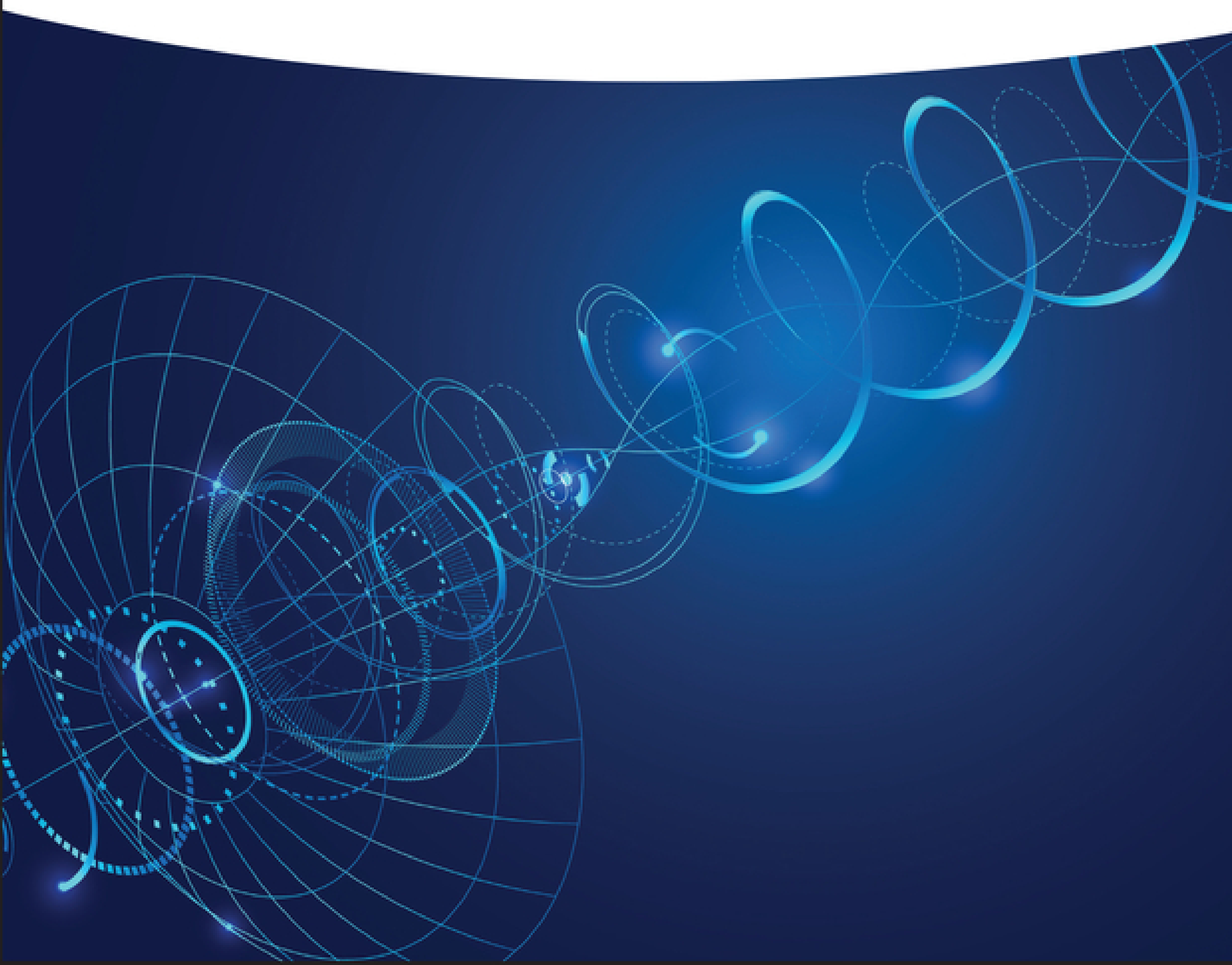

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Contents

Preface

With the rapid development of information technologies, the increasing frequency of electromagnetic waves transmission and reception has placed higher demand on the wave-transparent performances, mechanical properties, and environmental resistances of the composites in service. Polymer matrix wave-transparent composites present lightweight, high strength, low dielectric constant (ε) and dielectric loss ($\tan\delta$), and materials/structure/function integration, which have promising applications in the fields of aviation/aerospace, transportation, and 5G communications. In recent years, polymer matrix wave-transparent composites have become one of the key materials in many major engineering fields.

At present, the research progress and achievements in the field of polymer matrix wave-transparent composites are complex, and it is necessary to summarize the basic concept and research progress in this field for the reference of researchers. Currently, there are few books on polymer matrix wave-transparent composites. The authors wrote this monograph to summarize the latest scientific research achievements worldwide.

This book is divided into seven chapters. After the brief introduction to polymer matrix wave-transparent composites, it first introduces the measurement method of the dielectric properties as well as the mechanism and models of the wave transmission. Then, starting from reinforced fibers and polymer matrix widely utilized in recent years, it elaborates on the structures and properties of the common reinforced fibers and polymer matrix. Furthermore, the interface between the reinforced fibers and polymer matrix is also introduced. Finally, the practical applications of the polymer matrix wave-transparent composites are introduced in detail. Chapter 1 is written by Prof. Junwei Gu, where the introduction of polymer matrix wave-transparent composites is given from the basic concept, the composition of polymer matrix and reinforced fibers and their advantages and disadvantages, the factors influencing the wave-transparent performances, and the performance requirements under actual service condition. Chapter 2 is written by Dr. Jing Dang, Dr. Lin Tang, and Prof. Junwei Gu, where the relationship between the wave-transparent performances and ε and $\tan\delta$ of wave-transparent materials is discussed, and the equation for dielectric parameters, test methods for dielectric properties, and wave-transparent models are introduced. Chapter 3 is written by

Prof. Jie Kong, Dr. Zheng Liu, Dr. Jing Dang, and Prof. Junwei Gu, where the basic structure, physical and chemical properties, and modification methods of the polymer matrix are described in detail. Chapter 4 is written by Dr. Lin Tang, Prof. Yusheng Tang, Jiani Zhang, and Prof. Junwei Gu and mainly covers the structures, composition, preparation methods, and physical and chemical properties of the inorganic fibers (glass fibers and quartz fibers) and organic fibers (aramid fibers, ultrahigh molecular weight polyethylene fibers, and poly-*p*-phenylene benzobisoxazole fibers). Chapter 5 is written by Dr. Zheng Liu, Prof. Jie Kong, Dr. Jing Dang, and Prof. Junwei Gu, which describes the basic concepts of interfaces for polymer matrix wave-transparent composites, the formation of the interfaces, the mechanism of interfacial action, the interfacial characterization methods, and so on. This chapter also introduces the research progress of surface functionalization for reinforced fibers and the design and synthesis of interfacial compatibilizers. Chapter 6 is written by Prof. Yusheng Tang, Dr. Lin Tang, Yuhan Lin, and Prof. Junwei Gu, which introduces the structural design and molding principle of the polymer matrix wave-transparent composites as well as the processes of hand paste molding, pressure bag molding, laminated molding, resin transfer molding (RTM), winding molding, and so on. Chapter 7 is written by Dr. Jing Dang and Prof. Junwei Gu and introduces the application of the polymer matrix wave-transparent composites in radomes for aircraft, airborne, shipborne, ground vehicle radar,5G communication, printed circuit boards, and so on.

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On the occasion of the publication of this book, I would like to express my sincere thanks to the family, friends, colleagues, and editors of Wiley who have worked so hard to make the book go smoothly. We would like to thank the "Structure/Function Polymer Composites" (SFPC) research group of Northwestern Polytechnical University for their help in preparing this book. This book is mainly aimed at scholars, scientific researchers in universities and research institutes, as well as related technology developers of companies engaged in the research and development of polymer matrix wave-transparent composites.

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1

Introduction

1.1 Outline on Wave-Transparent Composites

Wave-transparent composites are a class of functional composites that can pass through electromagnetic waves. On the one hand, wave-transparent composites can provide electromagnetic windows for the transmission and reception of electromagnetic waves to ensure their efficient operation [1]. On the other hand, they can protect the radar antennas, communication, and microwave systems from the harsh external environment such as heavy rain, strong winds, snow, sand, solar radiation, and salt spray [2], ensuring the stability and reliability of electromagnetic wave transmission. With the rapid development of modern electronic information technology as well as the aviation and aerospace industries, the requirements for comprehensive performance of wave-transparent composites are becoming more and more demanding [3].

As far as matrix classification, wave-transparent composites can be divided into ceramic-based and polymer matrix wave-transparent composites [4]. Ceramic-based wave-transparent composites can meet the electrical performance requirements of radar radomes in the centimeter-band electromagnetic wave range. However, for millimeter-band electromagnetic waves (wavelength in the range of 1–10 mm and frequency in the range of 30–300 GHz), ceramic-based wave-transparent composites have disadvantages such as low strength, thick cover walls, and poor wave-transparent performances, which make it difficult to meet the performance requirements of radar radomes for millimeter wave [5, 6].

Polymer matrix wave-transparent composites have the advantages of lightweight, high strength, low dielectric constant (ε) and dielectric loss ($\tan\delta$), and materials/structure/function integration, which have a wide range of promising applications in satellite antennas, aircraft, missiles, 5G ground communication base stations, printed circuit boards, and so on. (Figure 1.1) [7].

This book will describe the wave-transparent mechanism, polymer matrix and reinforced fibers, their two-phase interfaces, molding process, and application prospects of the polymer matrix wave-transparent composites.

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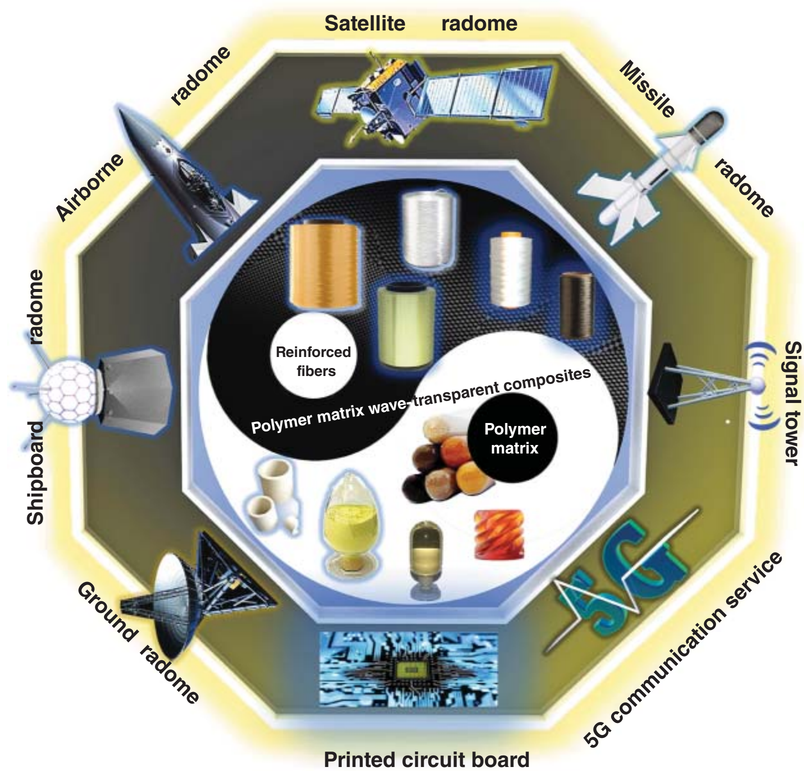

Figure 1.1 Application examples of polymer matrix wave-transparent composites. Source: Polymer matrix wave-transparent composites: A review. Journal of Materials Science & Technology, 2021, 75: 225–251 (Figure 1).

1.2 Composition of Polymer Matrix Wave-Transparent Composites

Polymer matrix wave-transparent composites consist of polymer matrix, reinforced fibers, and two-phase interfaces [8]. Polymers with low ε and $\tan\delta$ values as the matrix fibers with high strength and modulus as reinforced fibers produce advanced polymer-based composites (Figure 1.2) with both mechanical properties and wave-transparent performances *via* hot pressing, vacuum bagging, or resin transfer molding [9].

The heat resistance of polymer matrix determines the thermal stability of the composites in this case, and the fibers mainly serve as reinforcement [10]. Because the dielectric properties of different polymer matrices differ substantially. However, the ε value of reinforced fibers is generally larger than that of polymer matrix. Therefore, the selectively reinforced fibers possess excellent mechanical and thermal properties but also wonderful dielectric properties [11].

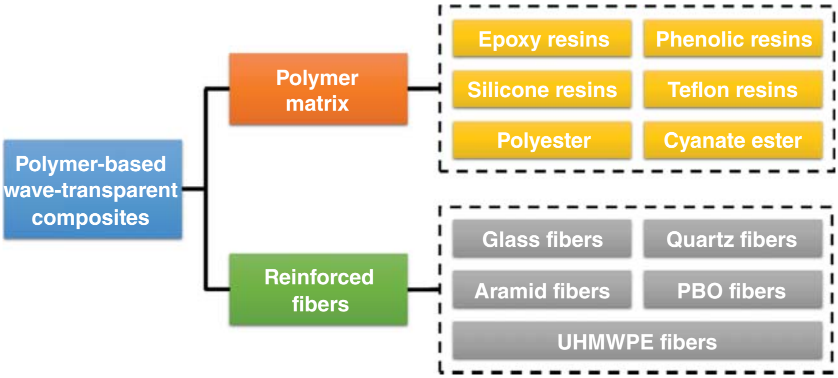

Figure 1.2 Composition of polymer matrix wave-transparent composites (commonly used polymer matrix and reinforced fibers).

1.2.1 Polymer Matrix

Polymers commonly used in wave-transparent composites mainly include epoxy resins [12], phenolic (PF) resins [13], polyimide (PI) resins [14], bismaleimide (BMI) resins [15], silicone resins, polytetrafluoroethylene (PTFE) resins [16], unsaturated polyester (UP) resins [17–19], and cyanate (CE) resins [20]. Table 1.1 shows the main physical and chemical properties of the common polymer matrix.

Epoxy resins have good flowability, low curing shrinkage, and high thermal decomposition temperatures (300–350 °C), but their high ε and $\tan\delta$ values limit their application in high-performance polymer matrix wave-transparent composites [21–23]. PF resins have good heat resistance (long-term service temperature at 250 °C), mechanical properties, and weatherability [24]. However, the ε values of PF resins increase significantly with increasing temperatures [25–27]. PI resins have high heat resistance ($T_g \geq 250$ °C), ε, and $\tan\delta$ values that remain stable over a wide

Table 1.1 Main physical and chemical properties of the common polymer matrix.

Types	Density (g/cm^3)	Flexural strength (MPa)	Flexural modulus (GPa)	ε (10^6 Hz)	$\tan\delta$ (10^6 Hz)
Epoxy	1.30	97	3.8	3.0	0.020
PF	1.30	92	3.5	3.2	0.020
PI	1.36–1.43	170	3.8	3.2	0.007
BMI	1.30	150	3.7	3.0	0.014
Organic silicon	—	85	—	3.0–5.0	0.003–0.050
PTFE	2.20	90	—	2.1–2.3	0.0003–0.0004
UP	1.29	85	3.2	3.0	0.018
CE	1.29	80	2.8	2.8–3.2	0.002–0.008

range of temperatures and frequencies [28]. At the same time, PI resins have excellent mechanical properties, chemical resistance, and dimensional stability [29–31]. However, PI resins are costly and difficult to process [32, 33]. BMI resins are an ideal polymer matrix for advanced composites due to their good heat resistance, excellent mechanical properties, relatively low ε value, resistance to humidity, chemical reagents, and good processability [34, 35]. However, the relatively high $\tan\delta$ values of BMI resins limit their wider application to a certain extent [36–38]. Silicone resins have excellent heat resistance and stable ε and $\tan\delta$ values under a wide range of environmental conditions [39–41], but their poor mechanical strength makes them rarely used alone [42–44]. PTFE resins have the lowest ε and $\tan\delta$ [45, 46] but are not easy to process and have low bonding properties between PTFE matrix and reinforcements [47–49]. UP resins have better mechanical properties than PF resins and have low ε and $\tan\delta$ values [50–52], which can be cured at room temperature. UP resins have a simple molding process, making them suitable for large-scale or large radome production [53–55]. However, UP resins have a short storage period, relatively low heat deflection temperature, and large curing shrinkage, which makes them unsuitable for the preparation of polymer matrix wave-transparent composites with high dimensional accuracy requirements [56–58].

In comparison, CE resins combine the high-temperature resistance of BMI and PI resins with the good processing properties of epoxy resins [59–61]. The highly symmetrical triazine ring structure and low polarity of the cured CE resins also make them low ε (2.8–3.2) [62–64], good heat resistance, and dimensional stability over a wide temperature and frequency range [65]. The structure and properties of commonly used polymer matrix are described in detail in Chapter 3.

1.2.2 Reinforced Fibers

Reinforced fibers for polymer matrix wave-transparent composites mainly include glass fibers [66, 67], quartz fibers [68], Kevlar fibers [69, 70], ultra-high-molecular-weight polyethylene (UHMWPE) fibers [71, 72], and poly(*p*-phenylene-2,6-benzobisoxazole) (PBO) fibers [73, 74]. Their main physical and chemical properties are shown in Table 1.2.

Table 1.2 Main physical and chemical properties of common reinforced fibers.

Properties Types	Density (g cm^{-3})	Tensile strength (GPa)	Modulus (GPa)	ε (10^6 Hz)	$\tan\delta$ (10^6 Hz)
E-glass fibers	2.54	3.75	72	6.13	0.0038
S-glass fibers	2.49	4.00	85	5.21	0.0068
D-glass fibers	2.6	2.40	52	4.00	0.0025
Quartz fibers	2.20	1.70	72	3.78	0.0002
Kevlar49 fibers	1.45	3.45	137	3.85	0.0010
UHMWPE fibers	0.97	5.01	193	2.25	0.0002
PBO fibers	1.56	5.80	280	3.00	0.0010

Figure 1.3 Application examples of D-glass fibers reinforced polymer matrix wave-transparent composites: MIRAGE 2000 (France, a); GRIPEN JAS 39 – Credit: Thierry ducros/Airliners.net (Sweden, b); HAWK 200 – Credit: Ben Stacey/Flickr (United Kingdom, c); HARRIER – Credit: Weimeng/Air Team Images (United Kingdom, d). HARRIER GR.9 – Credit: Titan Miller/Airliners.net. Source: (b) Ben Stacey/Flickr.

Glass fibers are the most commonly used inorganic reinforced fibers for wave-transparent composites. The earliest glass fibers used were E-glass fibers [75, 76]. Then, high-strength glass fibers (S-glass fibers) [77–79] and high-silica glass fibers (D-glass fibers) [80, 81] were developed to meet the special needs of aviation, aerospace, military, and other high-tech fields. Compared to E-glass and S-glass fibers, D-glass fibers have relatively lower ε and $\tan\delta$, which has been used in the radomes of MIRAGE 2000 (France), GRIPEN JAS 39 (Sweden), HAWK 200 (United Kingdom), and HARRIER (United Kingdom) (Figure 1.3) [82, 83].

However, with the rapid development of information technology, electronic components receive and transmit electromagnetic waves at increasingly high frequencies [84]. The high content of alkali metal oxides in glass fibers and the strong signal hysteresis and attenuation produced during electromagnetic wave transmission limit their application in high-frequency and high-precision wave-transparent composites [85]. Quartz fibers contain only a single component of silicon dioxide (SiO_2) with purity of over 99.9% and have excellent high-temperature resistance, electrical insulation properties and ablation resistance, low ε and $\tan\delta$ values, and so on [86, 87], which have been one of the most commonly used reinforced fibers in wave-transparent composites in the military and civilian sectors. However, quartz fibers have disadvantages such as high density, poor mechanical properties, and large ε values [88].

With the increasing demand for comprehensive performances of polymer matrix wave-transparent composites in terms of weight reduction, wave-transparency, and loading, researchers have carried out relevant research on organic reinforced fibers such as Kevlar fibers [89], UHMWPE fibers [90] and PBO fibers [91, 92]. Kevlar fibers, with low density, high specific strength, and specific modulus, are one of the most commonly used organic reinforced fibers in polymer matrix wave-transparent composites [93–95]. However, the high moisture absorption of Kevlar fibers is susceptible to moisture swelling and cracking, resulting in the degradation of wave-transparent performances and mechanical properties [96]. UHMWPE fibers, also known as high-strength, high-modulus polyethylene fibers, have a relative molecular mass of over 1 million, which is beneficial to outstanding impact resistance, cut resistance, chemical resistance and UV resistance, excellent low-temperature resistance, and low ε and $\tan\delta$ values [97, 98]. However, as the macromolecular chains of UHMWPE fibers are connected by a highly symmetrical methylene structure, the intermolecular Van der Waals forces are weak, making their T_g and melting point low, resulting in their high-temperature resistance and poor creep resistance [99].

Moreover, the surface of UHMWPE fibers does not contain polar groups, resulting in low surface energy, which creates poor bond strength between the UHMWPE fibers and polymer matrix [100, 101]. As a super fiber of the twenty-first century, the large number of rigid aromatic and oxazole rings in the PBO fiber molecular chain and a highly ordered crystal structure give PBO fibers excellent mechanical properties, heat resistance, chemical stability, and low ε (3.0) and $\tan\delta$ (0.001) values, which are of wide interest in the field of airborne/starborne radar radomes. Furthermore, PBO fibers have higher tensile strength, lower density, and ε values than those of inorganic reinforced fibers such as quartz [102]. Compared to those of other organic fibers, PBO fibers have about twice the strength and modulus of para-Kevlar fibers, and the thermal decomposition temperature of PBO fibers in the air is about 650 °C, which is approximately 100 °C higher than that of Kevlar fibers and much better than that of UHMWPE fibers (300 °C) [103]. As a result, PBO fibers have received a lot of attention as potential reinforcements for light weight/loading/wave-transparent integrated wave-transparent composites [104]. However, PBO fibers still have disadvantages of high cost, smooth and inert

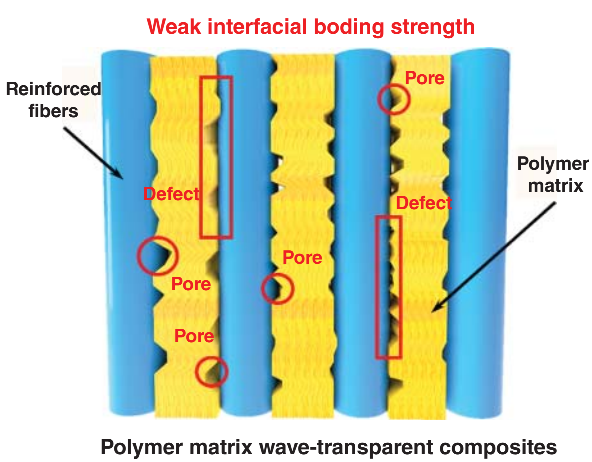

Figure 1.4 Schematic diagram of the two-phase interface for polymer matrix wave-transparent composites.

surfaces, and so on [105–107]. The structure and properties of these commonly used reinforced fibers are described in detail in Chapter 4.

In addition, the microscopic phase interface links the polymer matrix and reinforced fibers [108]. Defects are likely to arise at the poor two-phase interface (Figure 1.4), which would affect the overall performance (especially the interlaminar shear strength, ILSS) of the polymer-matrix wave-transparent composites [109, 110].

Therefore, how to effectively enhance the interfacial compatibility between polymer matrix and reinforced fibers has become a hot and difficult issue in this field [111]. Chapter 5 provides a detailed description of the two-phase interface inner polymer matrix wave-transparent composites and their optimal control strategies.

1.3 Factors Influencing the Wave-Transparent Performances of Polymer Matrix Wave-Transparent Composites

Polymer matrix wave-transparent composites are mainly used for electromagnetic windows and radomes in the fields of aviation/aerospace, 5G communication, and electronic information [112]. In order to ensure that all types of radar and antenna systems remain in stable operating conditions under harsh external environments, polymer matrix wave-transparent composites are required to have excellent wave-transparent performances (low ε and $\tan\delta$ values) [113].

The main factors affecting the wave-transparent performances of polymer matrix composites are divided into internal factors (intrinsic ε and $\tan\delta$) and external factors (thickness and electromagnetic wave frequency) [114, 115]. In general, the lower the molecular polarization rate and the density of polarized molecules of polymer matrix and reinforced fibers, the lower the ε and $\tan\delta$ of polymer matrix wave-transparent composites, the less energy is reflected and lost during the transmission of electromagnetic waves, and the correspondingly higher the wave-transparent rate [116]. In addition, polymer matrix wave-transparent composites are typically multiphase systems, and the interface between polymer matrix and reinforced fibers is prone to interfacial polarization, increasing the ε and $\tan\delta$ values, which is not conducive to improving the wave-transparent performance [117].

In addition, the thickness of polymer-based wave-transparent composites also affects their wave-transparent performances [118]. When the frequency of the electromagnetic wave is constant, the thickness of the wave-transparent composites increases, resulting in a tendency for the wave-transparent rate to decrease and then increase (Figure 1.5) [119]. This is mainly due to the reflection and loss (both absorption and interference shifts) that occur on the surface and inside the wave-transparent composites as the electromagnetic waves pass through [120, 121]. When the thickness approaches an odd multiple ($d = n\lambda/4$, $n = 1, 3, 5$, etc.) of its quarter wavelength ($\lambda/4$, Eq. 1.1), electromagnetic waves cause strong interference cancellation in the wave-transparent composites. This leads to an attenuation of the

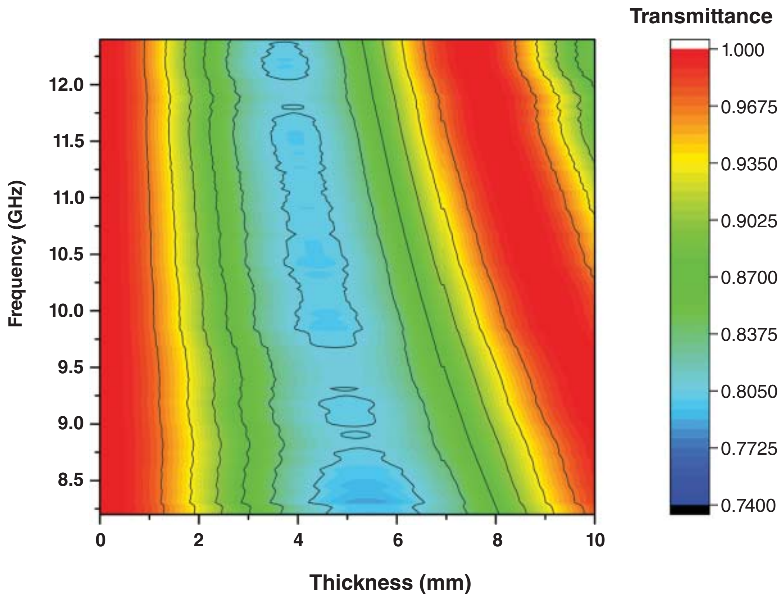

Figure 1.5 Schematic representation of wave transmission versus material thickness for polymer matrix wave-transparent composites.

electromagnetic wave energy and a significant reduction of the transmitted waves, resulting in the reduction of the wave transmission [122–124]. When the thickness is close to an even multiple ($d = n\lambda/4$, $n = 2, 4, 6$, etc.) of $\lambda/4$, the electromagnetic waves reflect less at the incident interface and can enter the interior almost unharmed, with the high wave transmission rate [125, 126].

$$n\lambda/4 = nc/4f_m * (u_r * \varepsilon_r)^{1/2} \tag{1.1}$$

where λ represents the wavelength of the incident waves; c represents the speed of light; f_m represents the frequency of the incident waves; u_r represents the magnetic permeability of the medium; and ε_r represents the dielectric constant of the medium.

1.4 Property Requirements for Polymer Matrix Wave-Transparent Composites

1.4.1 Wave-Transparent Performances

The ε and $\tan\delta$ values of polymer matrix wave-transparent composites are among the most important parameters affecting the wave-transparent performances [127]. In practice, the transmission rate of electromagnetic waves is usually required to exceed 70% in the broad frequency range (0.3–300 GHz), which usually requires the corresponding ε of polymer matrix wave-transparent composites to be stable in the range of 1–4 and $\tan\delta$ in the range of 10^{-2}–10^{-3}. Meanwhile, the ε and $\tan\delta$ are required to remain constant in the broad frequency and temperature range (0–220 °C) [128, 129].

1.4.2 Mechanical Properties

As structural loading materials, polymer matrix wave-transparent composites must have a certain degree of stiffness and strength to ensure the stability and reliability of the antenna system in various complex operating environments [130]. The tensile strength of polymer matrix wave-transparent composites for high-performance radomes is generally not less than 400 MPa. The compressive strength is more than 350 MPa to ensure the integrity of the antenna system under aerodynamic loads and impact of foreign objects, thus ensuring the normal operation of the electronic components inside the radomes [131–133].

1.4.3 Heat Resistant Properties

When the vehicle is flying at ultra-high speed in the atmosphere, the surface temperature of vehicle rises sharply with the increase in Mach number due to the heating of the high-temperature compressed gas between the excitation wave and the body and the strong friction between the surface of the body and the air (usually when the Mach number is 2, the surface temperature of the vehicle is about 150 °C; while when the Mach number increases to 3, the surface temperature rises sharply to about 350 °C, even exceeding the strength limit temperature of aluminum alloy) [134], therefore, when polymer matrix wave-transparent composites are used as radomes for aircraft, they should have excellent heat resistance (pyrolysis temperature greater than 300 °C) to overcome the high thermal stresses of external aerodynamic heating and to avoid deformation or even cracks under rapid temperature change [135, 136].

1.4.4 Environmental Resistance Properties

As protective materials for radar antenna systems, polymer matrix wave-transparent composites are subject to surface aging, polymer matrix degradation, and interfacial debonding between the polymer matrix and reinforced fibers during long-term service, which would seriously affect their service stability and reliability [137, 138]. Therefore, polymer matrix wave-transparent composites are required to have excellent environmental aging resistances. Current research revealed that environmental factors (humidity, heat, high and low-temperature alternation, and light) had a significant effect on the mechanical and dielectric properties of glass fiber-reinforced epoxy resin wave-transparent composites. When the relative humidity increased from 25% to 85%, the ε and $\tan\delta$ increased by 10% and 18.6%, respectively. In addition, the mechanical properties were strongly influenced by the hygrothermal conditions. The retention of tensile and flexural strengths after boiling for 200 hours was about 90%, but the retention of ILSS was only 61% [139].

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