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High-frequency welding of glass–fibre-reinforced polypropylene with a thermoplastic adhesive layer: Effects of ceramic type and long-term exposure on lap shear strength

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ABSTRACT

In this study, high-frequency (HF) welding of glass–fibre-reinforced polypropylene (GF/PP) with thermoplastic adhesive layers consisting of zinc oxide (ZnO), anatase-type titanium oxide or silicon carbide was investigated. Effects of the ceramic type and content on the dielectric and temperature characteristics of these adhesive layers were evaluated experimentally, and the ratio of the dielectric loss tangent to the relative dielectric permittivity ($\tan\delta/\varepsilon'$), which was the index of the HF heating efficiency, exhibited different tendencies by these parameters. This value increased rapidly with increasing temperature even at 10 vol% ZnO, suggesting that the heating of the adhesive layer may be accelerated by combining temperature rise with a small amount of ZnO. During the HF welding process, the ZnO-containing adhesive layer bound to GF/PP in the shortest time (18 s) with high bond strength (~14 MPa). The effects of temperature (50 °C) and moisture (80% relative humidity) on the mechanical strength of the HF-welded specimens after a long-term exposure were also examined.

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1. Introduction

Fibre-reinforced thermoplastics are becoming great interest for the industry because it enables rapid production and it is a good material choice for recycling. A variety of thermoplastics resins have been investigated as matrices, including polypropylene, nylon, polyetherimide, poly(phenylene sulphide), poly(ether ether ketone), etc. [1]. Among these resins, polypropylene (PP) is much attractive for the automotive industry, since it is light weight and inexpensive [2]. However, its poor bondability, owing to the low surface energy and inert chemical nature, has so far limited the widespread use of PP under mass production conditions where joining is necessary [3]. Therefore, several methods such as mechanical fastening, adhesive bonding combined with pretreatment (such as corona discharge, flame, fluorination, low-pressure vacuum plasma, atmospheric plasma and UV photografting) [3–6] and welding [7–13] have been applied to join PP or its composites. High-frequency (HF) welding is one of the welding methods to join plastics, e.g. polyvinyl chloride [14,15]. However, this technique cannot be directly applied to PP because its low loss factor leads to poor heat generation on HF irradiation [15]. In our previous study, the HF welding of PP by using thermoplastic adhesive layers such as silicon carbide (SiC), zinc oxide (ZnO) and anatase-titanium oxide (TiO_2) has achieved strong bonding and adherend failure in lap shear specimens [16]. Furthermore, this technique has been extended to the welding of glass–fibre-reinforced polypropylene (GF/PP), which exhibits higher strength than PP, by using a SiC-containing thermoplastic adhesive layer, and the effects of SiC content and particle size on welding properties have been investigated [17]. Consequently, the GF/PP was welded in a short time when high concentrations of small SiC particles were incorporated in the adhesive layer and the welded specimen exhibited a maximum lap shear strength of ~10 MPa.

In this study, the thermoplastic adhesive layers contained ZnO or anatase-TiO_2, which can be utilised for the HF welding of PP [16]. The influence of the ceramic type and content on the dielectric and temperature behaviour of the adhesive layer were examined and compared with previous results involving SiC. In addition, the HF welding of GF/PP was conducted using these adhesive layers, and the influence of ceramic type and content on welding time and bond strength were also investigated. The long-term effects of

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temperature and moisture on the lap shear strength of HF-welded specimens by using these thermoplastic adhesive layers were investigated.

2. Materials and methods

2.1. Materials

The GF/PP plate (as an adherend) and PP (as a base material for the thermoplastic adhesive layer) used in this experiment were selected the same materials with the Ref. [17]. Zinc oxide (Wako Pure Chemical Industries, Ltd., Japan, $\rho=5.6$ g/cm^3), anatase-TiO_2 (Kanto Chemical Co., Inc., Japan, $\rho=4.2$ g/cm^3) and SiC (GMF15H, Pacific Rundum Co., Ltd., Japan, $\rho=3.2$ g/cm^3) were mixed into the thermoplastic adhesive layer as ceramics heated by HF irradiation. For the same volume of SiC in the adhesive layer, GF/PP was quickly welded with high bond strength when the SiC particle size amounted to 0.54 μm [17]. Zinc oxide and anatase-TiO_2 particles were therefore selected near this size in this study. Particle size distributions were measured using a laser diffraction particle size analyser (SALD-3100, Shimadzu, Japan), and median sizes of diameter equalled 0.96, 0.64 and 0.54 μm for ZnO, anatase-TiO_2 and SiC, respectively.

2.2. Dielectric properties of thermoplastic adhesive layers

Thermoplastic adhesive layers involved in the ZnO, anatase-TiO_2 or SiC were prepared and measured their relative dielectric permittivity (ε') and dielectric loss tangent ($\tan\delta$) at a frequency of 40 MHz by the method written in the previous report [17]. The heating efficiency of a thermoplastic adhesive layer has previously been shown to depend on the $\tan\delta/\varepsilon'$ ratio of the layer [16,17]. Therefore, this ratio was calculated from the measured ε' and $\tan\delta$ values.

2.3. HF welding experiment and lap shear test of welded specimens

HF welding experiments were performed through the similar process as our previous work [17], and following specific conditions, which were optimised at the work, were used in this experiment. The thermoplastic adhesive layer was inserted between GF/PP plates and clamped at 0.03 MPa by the electrodes of the dielectric heating device (FDA-102PJ-01, Fuji Electronic Industrial Co., Ltd., Japan). Then, HF of 40 MHz was irradiated at an anode voltage of 3.5 kV and a current value of 130 mA. After melting the adhesive layer and welding the specimen, the HF irradiation was stopped and the clamping pressure was increased to 0.19 MPa and maintained at this level for 10 s to solidify the joint. Subsequently, the clamping pressure was released and the specimen was demoulded. Five replicate specimens were prepared for each combination of parameters considered.

This study aims to investigate the influence of the ceramic type on the bonding strength of GF/PP through a comparative evaluation. This evaluation was conducted using the single-lap shear joint strength, which was the simplest standard test used previously [17]. The lap shear strength was calculated by dividing the maximum tensile force by the welding area. The adhesive layer thickness was determined by measuring the thickness of welding area by using a micrometre (MDC-25MJ, Mitutoyo Corp., Japan) and subtracting that of two GF/PP pieces.

2.4. Tensile strength test of thermoplastic adhesive layer

Sheets of 2-mm-thick thermoplastic adhesive layers (prepared in Section 2.2) were cut into specific forms (Fig. 1) using a specimen cutting blade (Kobunshi keiki Co., Ltd., Japan). Using these specimens, tensile strength tests were carried out following the JIS (Japanese industrial standards) K7161:1994 standard with a testing machine (AG-100KNI, Shimadzu, Japan) at a crosshead speed of 2 mm/min.

2.5. Temperature and moisture effects on the lap shear strength

Temperature and moisture effects on the lap shear strength were evaluated by the method of Pint et al. [18]. Five replicate specimens were prepared for each combination of parameters. Single-lap specimens were prepared by HF welding using adhesive layers comprising either 10, 20 and 30 vol% ZnO, 10, 20 and 30 vol% anatase-TiO_2 or 20, 30 and 40 vol% SiC. Welded specimens were exposed to 50 °C and 80% relative humidity in a constant temperature/humidity chamber (SSE-24TR-A, Kato Inc., Japan) for 500 and 1000 h. The lap shear test was performed under conditions described in Section 2.3.

3. Results and discussions

3.1. Dielectric properties of thermoplastic adhesive layers

3.1.1. Dielectric properties of SiC-containing adhesive layers

Fig. 2 shows the temperature dependence of the dielectric properties of adhesive layers containing 10, 20, 30 and 40 vol% of SiC. The relative dielectric permittivities increased with SiC content, but no temperature dependence was observed. The dielectric loss tangent increased with increasing SiC volume and temperature. As a result, $\tan\delta/\varepsilon'$ became larger when the temperature and content of SiC increased, consistent with our previous study [17]. The increases in the dielectric permittivities with increasing ceramic content were reported in the $Sr_2Al_2SiO_7$ filled polyethylene composite [19], $Sr_9Ce_2Ti_{12}O_{36}$ filled polyethylene composite [20] and TeO_2 filled polytetrafluoroethylene composite [21]. In these reports, several theoretical models were introduced for predicting the relative dielectric permittivitiy of composites, for example, Jayasundare–Smith equation, Maxwell–Wagner equation, effective medium theory equation, Lichtenecker equation and Maxwell–Garnet equation. All of the equation indicated that the dielectric permittivities of the composites were increased with increasing the content of filler whose dielectric permittivities were higher than the matrix resin. We could not specify which equation fitted for our experimental result, because exact relative dielectric permittivity value of used powder-like SiC could not be measured.

3.1.2. Dielectric properties of ZnO-containing adhesive layer

Fig. 3 shows temperature effects on the dielectric properties of adhesive layers incorporating 10, 20, 30 and 40 vol% ZnO. The adhesive layer exhibited an increase in relative dielectric permittivity with increasing ZnO content, but no temperature dependence was visible (Fig. 3(a)). These trends were the same as that for SiC-containing adhesive layers, albeit with lower relative dielectric permittivities. The dielectric loss tangent increased with increasing temperature and slightly increased with ZnO content (Fig. 3(b)). Compared with adhesive layers presenting the same SiC

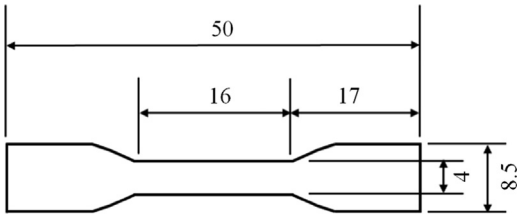

Fig. 1. Thermoplastic adhesive layer specimen for tensile test.

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