Development Of A Polymer Composite With High Electrical Conductivity And Improved Impact Strength For The Application As Bipolar Plate

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Abstract. Bipolar plates constitute the most important structural component in fuel cell stacks. Highly filled thermoplastic composites with high electrical conductivity obtain an increasing importance in the design of bipolar plates as alternative to conventional metallic systems. Thermoplastics (e.g. PP) have suitable properties such as a good processability, chemical resistance, light weight and low production costs. As thermoplastics have low electrical conductivities, conductive fillers have to be included in the matrix. A high content of such fillers (e.g. graphite) in excess of 80 wt.-% is necessary to achieve the desired electrical properties. However, materials with such high filler contents embrittle readily. The workability in injection and compression molding is difficult and the mechanical stability is insufficient in case of strain deformation. As consequence, material failure and an inacceptable amount of damaged goods can be observed during the processing.

As no suitable thermoplastic system is available for better mechanical properties, the induction and dispersion of a rubber phase in the thermoplastic matrix can be used to increase the impact strength of the conductive composite.

In this research work a ternary composite, based on PP as matrix, EPDM as impact modifier and synthetic graphite as conductive filler, was developed. The material was produced using a 26 mm co-rotating, intermeshing twin-screw extruder. The amounts of PP, EPDM and graphite were varied systematically and a process window was defined that enables improved impact strength and high electrical conductivity of the new material. The results indicate that impact strength can be enhanced by about 99 % with an EPDM content of 30 wt.-% in the PP matrix. The electrical conductivity decreases in a small range with increasing content of EPDM, but the conductivity is still excellent for producing bipolar plates.

1. INTRODUCTION

To reduce global CO_2 emissions, a change from fossil fuels to renewable energy sources is necessary. But energy production from renewable sources depends on weather, environment and time of day. Therefore, it is important to develop new technologies to store this energy and make it available again at later time. Two technologies that are currently to be researched are fuel cells and redox flow batteries[1-3].

A component, which is used in both of these technologies, is the bipolar plate. Bipolar plates have to distribute fuels or electrolyte solution on the reactive membrane. Bipolar plates are electrodes for the resulting voltage. Therefore, bipolar plates must exhibit a low electrical resistance, good thermal conductivity, mechanical strength, chemical resistance, and low gas permeability. Materials that are used for producing such bipolar plates are especially graphite, metal, and conductive filled plastic composites. However, each of these materials has disadvantages. Pure graphite is very difficult and costly to manage and it exhibits a low mechanical strength. Plastics are easy to process and often have got a good chemical resistance; however they are electrical insulators compared to metals and pure graphite.

To achieve surface resistances as they are required for bipolar plates, composites must be manufactured with very high degrees of filling. However, such composites exhibit very low impact strength and high brittleness due to the high filler content[4-5].

To increase the efficiency of bipolar plates made from highly filled plastic composites, the impact resistance of the composites has to be increased in accordance with the electrical and thermal conductivity. One way to improve the impact resistance of thermoplastics is mixing with a rubber[6].

The effect of a rubber component as impact modifier on the material properties of electrical and thermal conductive, highly filled plastic composites has not yet been examined in depth in research. For this reason, the material properties of impact-modified, conductive plastic composites are investigated in this work. These blends based on polypropylene and thermoplastic rubber filled with graphite are produced in a twin-screw extruder. The graphite content is varied to investigate the influence of material composition on the material properties and blends without and with 30 wt.-% rubber modifier is compared.

2. EXPERIMENTAL

2.1 Material

Polypropylene (PP) is characterized by good chemical resistance, good processability, low price and has been already used as a material for bipolar plates. In this work thermoplastic PP (SABIC PP 579S) of SABIC Germany GmbH, Düsseldorf, is used. An EPDM rubber (Keltan 2070P) of LANXESS Germany GmbH, Dormagen, is used as impact-modifier. In order to produce electrical and thermal conductivity of the composites, synthetic graphite powder (TIMREX® KS500) is applied with particle sizes in the range of 500 micrometer provided by Timcal Germany GmbH, Düsseldorf.

2.2 Processing and Analytics

A co-rotating, intermeshing twin screw extruder (ZSK26Mc) of Coperion GmbH, Stuttgart, Germany with a screw diameter D of 26 mm and a length of 44 D was used for compounding. Since the electrical and thermal conductivities depend strongly on the filler content, various graphite contents from 30 wt.-% up to 90 wt.-% were chosen. Weight percentages of 0 wt.-% and 30 wt.-% of EPDM within the PP matrix were used to investigate the effects of EPDM on the impact strength and the conductive properties. In addition, reference samples without EPDM were produced.

The materials were compression molded to plates with the dimensions 220 mm by 200 mm with a thickness of 4 mm in a heated press. From these plates samples for impact strength tests were prepared. The surface resistance of the pressed plates was measured with Loresta GP device of the company NH Instruments, Willich, Germany. The average value of 20 different measuring points for each sample was taken as the shown surface resistance. The thermal conductivity was determined at 25°C with the Nanoflash method in the thickness direction of the samples using the LFA 447 NanoFlash of NETZSCH-Gerätebau GmbH, Selb, Germany. The impact strength was measured by Charpy impact test in accordance with DIN EN ISO 179-1/1fU.

For optical analysis the samples were embedded in an epoxy resin, ground and polished. The samples were analyzed with a microscope from Keyence VHX - 700F Type of KEYENCE Germany GmbH, Neu-Isenburg in the reflected light mode.

3. RESULTS

Conventional graphite plates have low impact strength and are brittle. As illustrated in Figure 1a, the advantage of replacement of a part of PP by EPDM is clearly seen in the improvement of the impact strength in comparison to reference samples without EPDM. For composites containing 30 wt.-% graphite the impact strength could be enhanced from 5.50 kJ/m² to 8.35 kJ/m² by about 51 % with the addition of 30 wt.-% EPDM. At 90 wt.-% graphite content the effect decreases to an improvement of 44 %. The maximum value of the impact strength effect is reached at 60 wt.-% graphite with 99 % improvement.

Figure 1b shows the surface resistance for the samples with 80 wt.-% and 90 wt.-% of graphite. The samples with 30 wt.-% and 60 wt.-% graphite have to be classified as insulators. The surface resistance decreases significantly with increasing graphite content. Without EPDM, the resistance of 24.11 mΩ at 80 wt.-% graphite drops to 4.49 mΩ at 90 wt.-% graphite. The EPDM content of the matrix increases the surface resistance of the composites. The effect of EPDM on the surface resistance is larger at the higher graphite content. At 80 wt.-% and

90 wt.-% the surface resistance increases by about 31 % and 130 %, respectively, compared to the composites without EPDM. However, the value of 10 mΩ reached at 90 wt.-% graphite and 30 mΩ at 80 wt.-% graphite is in an acceptable range.

The graphite content has a very strong effect also on the thermal conductivity as shown in Figure 1c. Whereas at 30 wt.-% graphite a relatively low value of 0.6 W/(mK) was measured, the thermal conductivity increases to almost 40 times at the loading of 90 wt.-% graphite resulting in a value of 24.1 W/(mK). When adding EPDM, similar or slightly higher values are observed at 30 and 60 wt.-% graphite loading and a reduced value of 20 W/(mK) at a filling grade of 90 wt.-% graphite.

The optical micrograph in Figure 2 shows the morphology of the blend composites illustrating the graphite particles in the PP/EPDM matrix. It is seen that the particle diameter of the graphite is rather high despite the high shear stresses during the compounding process. The micrograph also illustrates a compact sample structure and a good wetting of graphite by the PP/EPDM matrix.

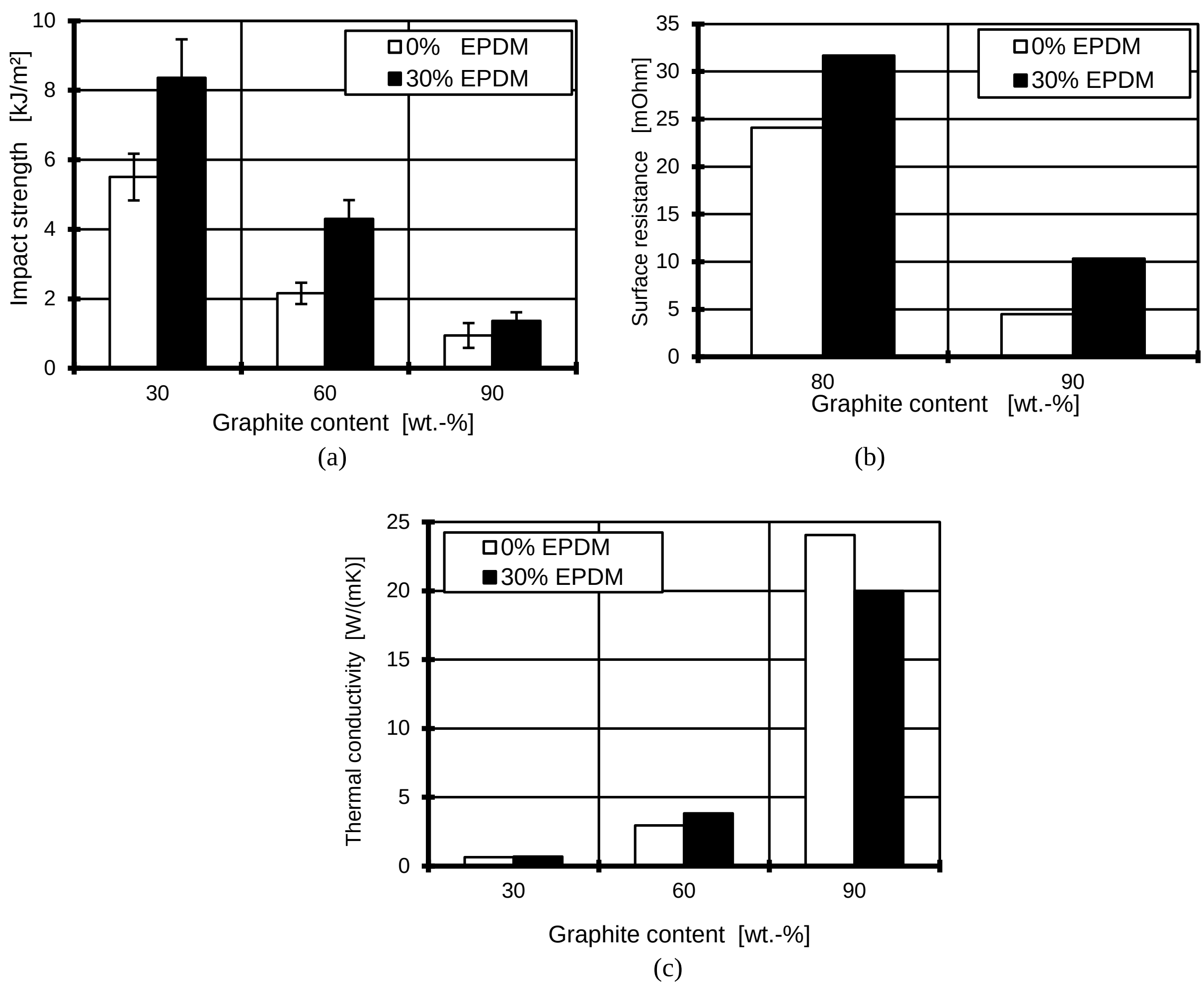

FIGURE 1. Impact strength (a), surface resistance (b), and thermal conductivity (c) of PP based bipolar plate materials dependent on the filler content and the use of EPDM as matrix modifier.

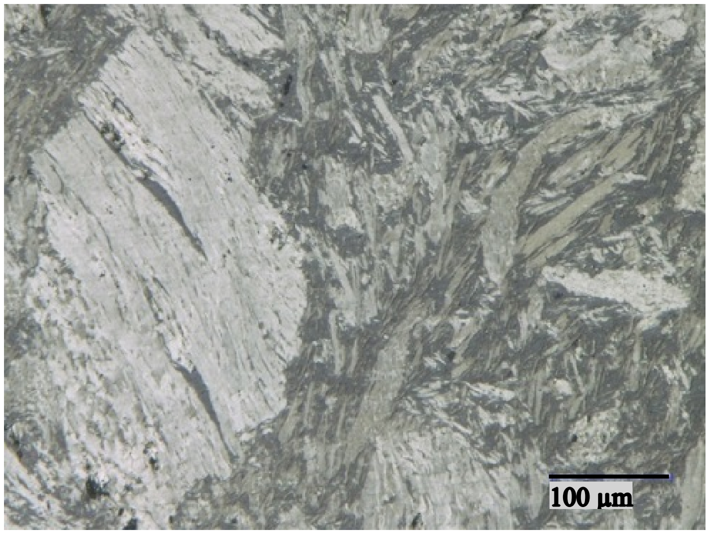

FIGURE 2. Optical micrograph of the PP based composite with 80 wt.-% graphite and 30 wt.-% EPDM in the PP matrix.

3. SUMMARY

Composite materials based on polypropylene and EPDM with different amounts of synthetic graphite have been produced using a laboratory scale compounding process. The properties surface resistance and impact strength are characterized, which are important for the application of such materials as bipolar plates. The impact strength and the surface resistance decrease with increasing graphite filler content. The decrease in impact strength can be partially compensated by the addition of 30 wt.-% EPDM which increases the impact strength up to 99 % at 60 wt.-% graphite loading. The thermal conductivity increases significantly up to 24 W/(mK) at 90 wt.-% graphite in the composite, whereas the EPDM addition decreases it slightly to 20 W/(mK). A minimum value of surface resistance of 4.49 mΩ is observed in the composite of 90 wt.-% graphite without EPDM. The addition of EPDM results in an increase of the surface resistance by about 31 % at 80 wt.-% graphite and by 130 % at 90 wt.-% graphite; however the values are still in an acceptable range.

The addition of EPDM within the PP matrix in a highly graphite filled composite leads to the desired improvement of the impact strength. The composite composition has to be further optimized; especially lower graphite filler contents for injection moulding have to be achieved without losing or reducing the conductive properties.

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