

H.-P. Heim¹, F. Mieth¹, A. Schlink^{1*}, K. Wiegel², L. Brabetz²

¹Institut für Werkstofftechnik/Kunststofftechnik, University of Kassel, Kassel, Germany

²Fachgebiet Fahrzeugsysteme und Grundlagen der Elektrotechnik, University of Kassel, Kassel, Germany

Joining of Contact Pins and Conductive Compounds via Injection Molding – Influence of the Flow Situation on the Electrical Contact Resistance

While a lot of research can be found in the field of bulk resistance of carbon filled polymers, comparatively few papers focus on contact resistance between compound and metal contacts. Due to that small number of researches that deal with contact resistances, studies of the influence of injection molding conditions and parameters on the contact resistance are also very rare. In contradiction to that, these influences on bulk resistance have been studied. The objective of this work was to investigate the electrical contact resistance of overmolded tinplated copper contacts after modifying flow situations and molding conditions by procedural and constructional methods in contact areas. Metal pins were overmolded with a polypropylene compound containing 45 vol.% graphite, utilising an insert injection molding process. To affect the flow situation at the contacts, several processing parameters, such as mold temperature and injection speed, were modified. In addition, the contact alignment related to melt flow direction was varied. Electrical properties were studied and related to macroscopic and microscopic connection properties and flow situations in contact regions. It was found that the contact resistance is a significant factor while examining electrical resistances of overmolded samples. Furthermore, it was shown that the various flow situations had an essential impact on contact resistances. Weld lines at the position of the contact caused a decreased contact resistance. The correlation of the weld line effect, the filler orientation and contact resistance were successfully investigated by μ -CT. Regarding processing parameters, it was observed that a high mold temperature of 120 °C increased not only bulk conductivity, but also had a positive impact on contact conductivity. Macroscopic and microscopic connection mechanisms of contact surfaces were interpreted and connected to the experimental observations.

* Mail address: André Schlink, Institut für Werkstofftechnik, Kunststofftechnik, Universität Kassel, Mönchebergstr. 3, D-34125 Kassel, Germany
E-mail: andre.schlink@uni-kassel.de

1 Introduction

Electrically conductive plastic compounds have a high potential for integrating functions in plastic parts. Depending on the level of their electrical resistance, they can be employed to avoid electrostatic charging or for shielding applications in power transmission. There are also special areas of application such as in electrical resistance heaters. To permit the industrial implementation of these applications, it is necessary to have knowledge of the electrical resistance of the materials when they are in service. This includes not only the bulk resistance but also the contact resistance that develops at the interface between the contact and the compound when electrical contacts are overmolded.

Bulk resistance has been under investigation for a long time and is a function inter alia of the distribution and orientation of the conductive fillers in the part, making it highly dependent on processing (Dörner, 2012; Wegryzn, 2013; Doagou-Rad et al., 2018). Descriptions are thus available of the way in which orientation influences conductivity, and it is also possible to simulate parts produced by injection molding, for example (Schneidmadel et al., 2016; Shokri and Bhatnagar, 2006; Li et al., 2008; Chen et al., 2018).

The fact that contact resistance is also highly dependent on processing is shown in what follows. The focus here is on the influence of the flow situation on the contact resistance of copper pins contacted via injection molding.

2 Current Status of Research

The prerequisite for electrical conduction processes in plastic compounds is the formation of a conductive filler network which creates conductive paths through the insulating matrix (Kirkpatrick, 1973; Zallen, 1998). Mathematical models of the formation of a conductive network have been established (Kormakov et al., 2019). Compounds with fillers that have an aspect ratio deviating greatly from 1 in particular are subject to process-conditioned orientation when injection molded.

Based on the shear rate profile, a multilayer model results over the flow cross-section. Different models define 3, 5 or 7 layers here (Papathanasiou, 1997). In all the models, fillers in the shear zone close to the mold wall are oriented parallel to the direction of flow, while those in the core zone are oriented randomly or perpendicular to the flow direction (SadAbadi and Ghasemi, 2007; Sun, 2015; Régnier, 2008).

Deviating from the ideal case of a continuous and homogeneous flow cross-section, this orientation can be modified through geometric constraints. The greatest influence is exerted by flow obstacles that divide the melt flow. When the flows merge, weld lines result. The intensity of these lines is determined inter alia by the angle at which the flows merge and by the temperature of the melt front (Dzulkipli and Azuddin, 2017). Even if the melt flows do not divide, however, filler distribution and orientation differing from the standard model can result at points where there is a change in cross-section (Schneidmadel et al., 2016). Since phenomena such as weld lines can result in especially pronounced differences in filler orientation, they are particularly suitable for examining the influence that the flow situation has on the electrical contact resistance.

When observing electrical contact resistances between two metal surfaces, approaches based on the model drawn up by Holm and Holm (1967, pr. 1979) are frequently adopted. A distinction is drawn between the apparent and the actual points of contact on the surface so that the surface area that is actually available for the current flow can be determined. Due to the small proportion of actual contact points (alpha spots), constriction resistance develops. Wabner (2001) transposed this model to plastic/metal contacts, but restricted this to contacting after injection molding. He does, however, take into account the fact that conduction is achieved through the filler contained in the compound and hence that the position of the filler in relation to the metal surface (distance, orientation) plays a decisive role.

Existing investigations into the contact resistance between a metal and a conductive compound in injection molding have shown that the contact resistance is lower than that for pressed-on electrodes (Hopmann and Haase, 2015). Investigations to date have identified the specific conductivity of the contact pin and the size of the contact surface as the factors with the greatest influence on the contact resistance between the overmolded contact pin and the plastic matrix. The position of the contact pin and the gate, and also any pre-treatment of the contact pin would appear to be of subordinate importance (Hopmann et al., 2013).

A detailed investigation of the influence of the flow situation on the electrical contact resistance is missing up to now and therefore should be addressed in this research.

3 Materials

The compound used for the investigations is a polypropylene (575P, Sabic, Riyadh, Saudi Arabia) which had 45 vol% graphite added to it on a twin-screw extruder (macro-crystalline natural graphite from Remacon, Bad Säckingen, Germany). The manufacturer specifies the particle size as 100% < 200 μm , 40% < 100 μm ; 20% < 70 μm . No drastic reduction in particle

size due to compounding or injection molding was observed in microscopic images. In isolated cases, particle sizes were identified that were even above the 200 μm limit specified by the manufacturer. The contact pins that were overmolded are countersunk head rivets according to DIN 661 (DIN 661: Countersunk head rivets – nominal diameters 1 mm to 8 mm) in bare copper. The dimensions are shown in Fig. 1.

Since oxide layers on electrical contacts impair their efficient connection to the electrically conductive material, the oxide layer must be removed from the contact pins and a metal coating applied that is suitable for electrical contacts and less prone to the formation of oxide layers. The copper rivets are thus given a tin coating. To do this, any copper(II) oxide on the copper rivets is removed through two hours' immersion in 25% acetic acid.

In the next step, the copper rivets which had had their oxide layer removed underwent chemical tin coating. Use was made here of Seno 3211, BESTCHEM GmbH, Linsengericht, Germany, tin powder for gloss tin baths. This resulted in a tin layer with a thickness of 2 μm .

4 Sample Preparation

The samples were produced on a plunger injection molding machine (model Haake Minijet II, Thermo Fisher Scientific, Waltham, USA). The injection mold used is a test specimen mold measuring $4.2 \times 12.5 \times 75.7 \text{ mm}^3$ which was equipped with mounts to hold pins. To achieve this, two holes were drilled in each mold half and two pin holders each inserted in them. The round shape of the pin holders means they can be rotated through 360° . The alignment of the pins can thus also be varied. The layout of the mold is described in Fig. 2.

After the four pin holders have been inserted into the two mold halves, two sets of pin holders are positioned opposite each other when the mold is closed. Once the mold has been closed, the top surfaces of the pin holders opposite each other are sealed, preventing the pin head from being wetted with melt and ensuring that it can subsequently be used for contacting. There is also a third insertion position at the end of the flow path. The result is then a test specimen with three overmolded contact pins, as shown in Fig. 3.

During the injection molding tests, the injection speed and the mold temperature on the injection molding machine were varied, in addition to the alignment of the pins. Since the injection speed could not be controlled directly, the injection pressure was set and, in preliminary tests, an average value for the injection speed determined from the filling time. The settings

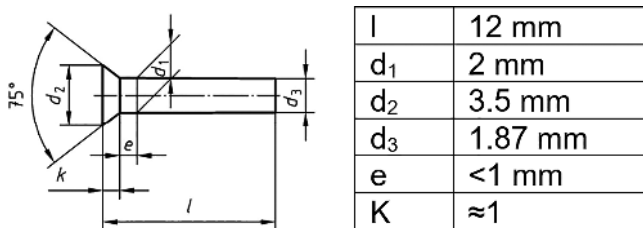


Fig. 1. Dimensions of the contact pin (shape as per DIN 661)

for the parameter variations can be seen in Table 1. The total quantity includes both alignments of the pins.

5 Electrical Measurements

All the measurements were performed using a standard four-wire method e.g. described in DIN EN ISO 3915. To avoid any influence due to the samples heating up, the maximum power input during measurement was less than 25 mW. Preliminary tests with a thermal camera showed that specimen heating due to the measuring current is less than 1 °C.

First of all, the total resistance between the overmolded pins was determined by applying clamps to them.

To exclude any influence of post-crystallisation processes, specimens were first measured immediately after demolding and then at different intervals up to 100 h (Fig. 4)

It was seen that, in the first 12 h after production, resistance falls by around 10 % and then remains at an approximately constant level. Storage of the specimens under defined conditions (23 °C, 45 % rH) for at least 96 h prior to measurement was thus specified as a condition.

Measurements were also conducted on partial areas of the sample. The sample was cut into individual parts to this end. It must be noted here that a 1 mm-wide section of the overall volume is lost due to the machining process. For contacting purposes, the resulting cut surfaces were coated with conductive silver lacquer ($v_{ag} = 47.25\%$) and dried for 12 h. Contacting was performed by pressing on a copper plate positioned in parallel to the contact surface with a contact pressure of 34.57 kPa for resistance $R_{comp.}$ and 45.5 kPa for resistance areas $R_{trans.-n}$ and $R_{trans.-f}$. The indices n (near) and f (far) describe the position of the pins in relation to the gating point. An overview of the individual areas is set out in Fig. 3. This

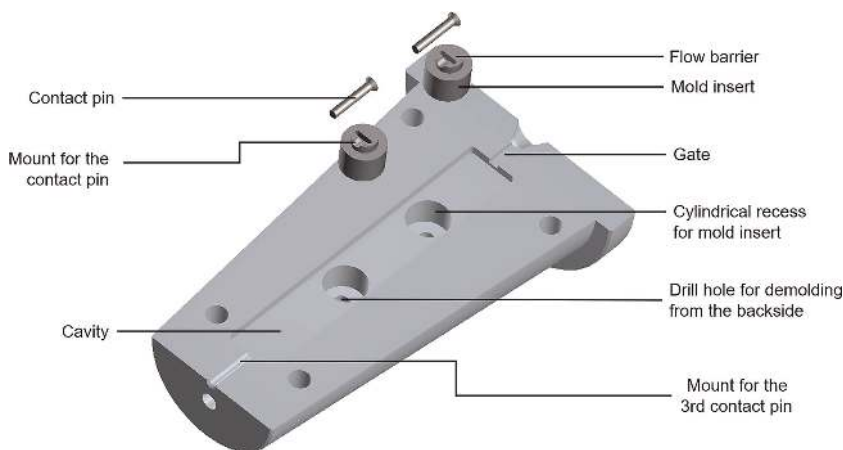


Fig. 2. Injection mold with inserts as flow barriers and pin fixtures

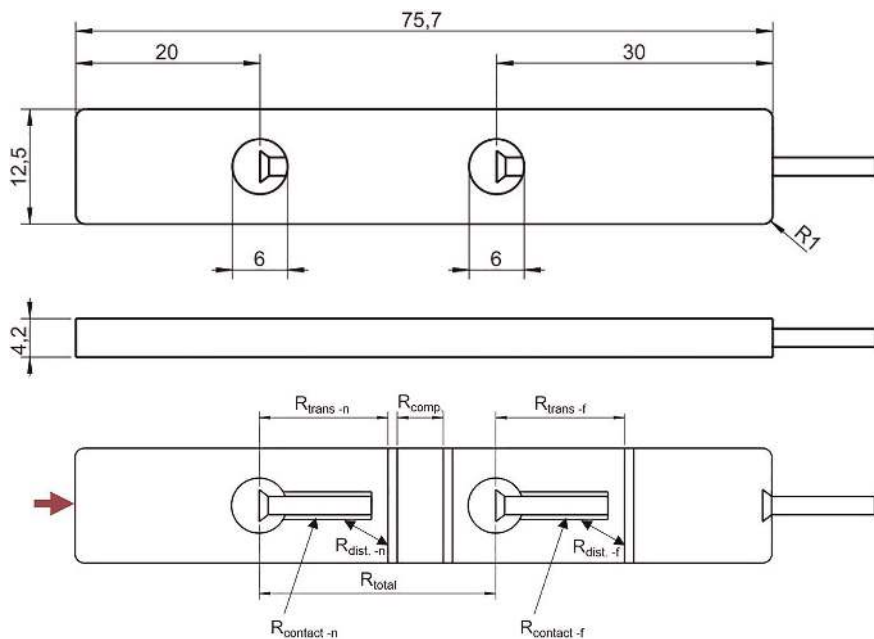


Fig. 3. Specimen with 3 contact pins and resistance areas

| Parameters | | Set 1 | Set 2 | Set 3 | Set 4 |
|----------------------------|------------|-------|-------|-------|-------|
| Injection pressure (bar) | P_{inj} | 120 | 120 | 500 | 500 |
| Packing pressure (bar) | P_{pack} | 100 | 100 | 450 | 450 |
| Flow front velocity (mm/s) | V_{inj} | 90 | 90 | 415 | 395 |
| Mold temperature (°C) | T_{mold} | 40 | 120 | 40 | 120 |
| Bulk temperature (°C) | T_{bulk} | 280 | 280 | 280 | 280 |
| Cooling time (s) | T_{cool} | 30 | 30 | 30 | 30 |
| Total quantity | | 10 | 10 | 10 | 10 |

Table 1. Injection molding parameters

shows the variant with the pins orientated in the direction of flow. Preparation of the samples with pins oriented against the direction of flow, i. e. pins that are rotated by 180° in the holder and have their tips pointed toward the gate, is performed in the same way.

In order to determine the interface resistance ($R_{contact}$), the samples required further mechanical processing. To do this, the interface layers are removed, the associated contact pin removed and the resulting hole in the plastic compound enlarged by drilling it to a diameter of 3 mm. The depth remains unchanged, however. This method does not scale with any variation in the interface thickness. The large diameter means that clearly more than the interface layer is removed, thus ensuring that the interface is indeed removed at all events. The material removed through drilling is shown in Fig. 5 in relation to the interface.

In the next step, the hole from which the interface has now been removed has conductive silver lacquer applied to it or is alternatively contacted with a tin-coated contact pin of the cor-

responding diameter, and the resistance determined using the method employed so far. The difference between the resistance measured in this way and the transfer resistance $R_{transfer}$ at this point, gives the interface resistance $R_{contact}$. An overview over the quantity of test specimens used for electrical measurements is shown in Table 2.

6 Results

Figure 6 shows the total resistance measured between the two overmolded contacts. It is clear that positioning the pins behind the flow obstacle and hence in the weld line led to lower total resistances. It was also seen that samples produced with higher mold temperatures had lower resistances. No clear influence is evident with respect with the injection pressure and the resulting flow front speed.

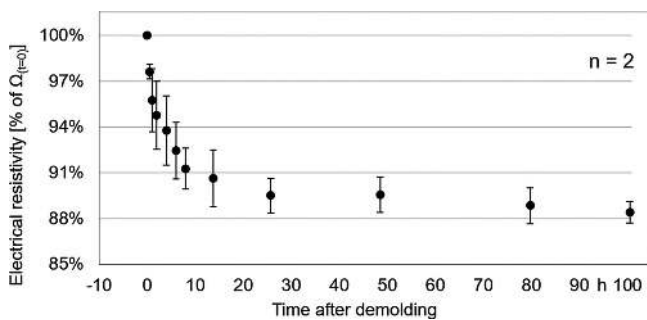


Fig. 4. Change in total resistance for multiple measurements of the specimens with an increasing storage time after injection molding

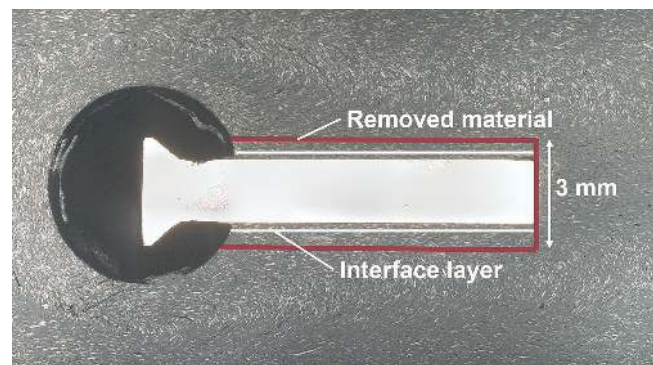


Fig. 5. Real interface area in comparison to the area removed by drilling

| Set | R_{total} | $R_{Contact -n}$ | $R_{Contact -f}$ | R_{Comp} | $R_{dist. -n}$ | $R_{dist. -f}$ |
|-----|-------------|------------------|------------------|------------|----------------|----------------|
| 1 | 10 | 4 | 4 | 4 | 4 | 4 |
| 2 | 10 | 4 | 4 | 4 | 4 | 4 |
| 3 | 10 | 4 | 4 | 4 | 4 | 4 |
| 4 | 10 | 4 | 4 | 4 | 4 | 4 |

Table 2. Quantities of electrical measurements per parameter set

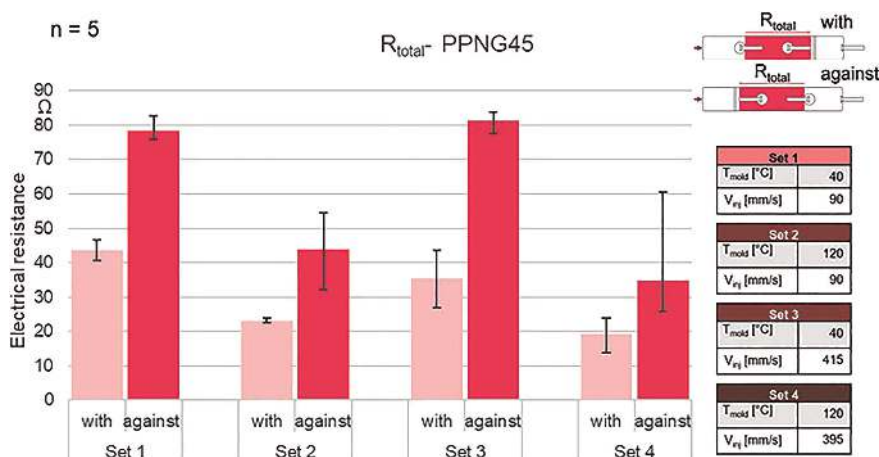


Fig. 6. Total resistance between the pins

For further understanding of how the total resistance is made up, it is possible to evaluate the shares of the overall resistance accounted for by the resistances of the individual segments, as shown in Fig. 7. This presentation includes all the set mold temperatures and injection pressures, since the ratio of the partial resistances to each other lies within a similar range. The actual line resistance of the compound, measured in an area approximately 5 mm long approximately parallel current flow, accounts for only a very small share of the total resistance, less than 10% in most cases. The values are in the region of 1.2 to 3.3 Ohms. With a cross-section area of 0.504 cm² and a conductor length of 0.5 cm, these correspond to the specific resistance in Ohm cm at the same time. The area between the contact pin and the area with a parallel current flow is defined as the transfer resistance R_{trans} . This is divided into the contact resistance directly at the pin $R_{contact}$ and the remaining transfer resistance R_{dist} . As mentioned before, indices n and f describe the pin position, where n stands for the pin inserted close to the gate and f for the pin further away from the gate. R_{dist} will later be divided in R_{dist_A} and R_{dist_B} . Figure 7 shows that, of the two resistances that are decisive for the transfer resistance, the direct contact resistance to the pin ($R_{contact}$) is the more important one. Together, the contact resistances $R_{contact-n}$ and $R_{contact-f}$ account for almost 70% of the total resistance of the samples on average. R_{comp} and R_{dist} both are conduction resis-

tances of the compound material and therefore depend on conduit length. Since the conduit lengths are comparable also R_{comp} and R_{dist} have values in the same range.

If just the contact resistance ($R_{contact}$) is observed in Fig. 8, a similar picture is seen for the influence of the pin orientation

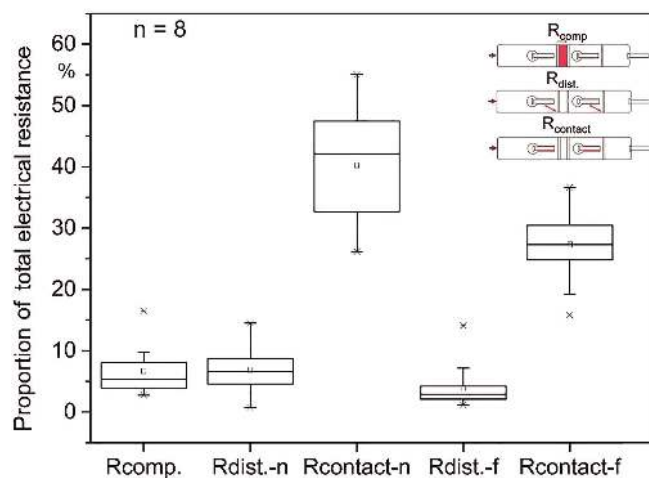


Fig. 7. Proportion of partial resistances in the total resistance between the pins

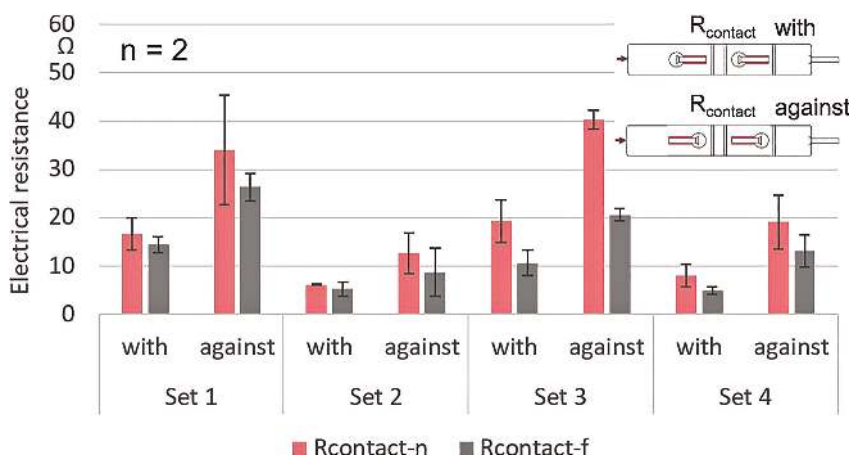


Fig. 8. Influence of processing parameters of contact resistance

and the processing parameters as with the total resistance. This behavior is also to be expected given the high share of the total resistance accounted for by the contact resistance.

7 Discussion

On average, approximately 70% of the samples' total resistance can be attributed to the two interface resistances. Consequently, the parameter influences that have been recognised must also be interpreted with respect to the interface resistances. To do this, it is first necessary to make clear, once again, how the interface resistance has been defined in this investigation. It covers an area 0.5 mm thick around the pin and thus comprises two parts – the resistance directly at the metal/plastic interface and the conduit resistance within the compound for conducting the current away from the boundary area. The latter is not, however, comparable with the line resistance R_{comp} , since, in the edge zone of the compound, a clearly different situation prevails in terms of filler orientation. Figure 9 contains a schematic diagram of the situation in the interface and the transfer area.

The study of different microscopic pictures showed that a separation formed between compound and pin in a macroscopic manner. Hence major interface areas are not in close contact (Fig. 9,1). Furthermore, microscopic gaps were observed. In that case, the two interfaces are in a macroscopic contact but not all microstructures are entirely covered by the compound (Fig. 9,2). Differences in filler concentration and orientation could be detected within the compound. The first layer starting at the interface is a polymer skin layer with lower filler concen-

tration, which is followed by a distribution layer (R_{dist_A}) with low orientation. Finally, a distribution layer with higher orientation (R_{dist_B}) leads to the transition to the regular compound conduction phase.

Based on this observation and existing knowledge about conductive networks in polymer compounds and electrical contact resistances for polymer and other materials, it is possible to define four conditions for a low contact resistance. Macroscopic contact must exist between the compound and the pin. The microstructure of the pin surface must be reproduced as accurately as possible by the melt. The fillers in the compound must be as close as possible to the surface or even protrude through it. The fillers closest to the interface must be well connected to the filler network.

7.1 Macroscopic Contact

As a rule, there will be macroscopic contact between the pin and the compound at the start of the packing pressure phase. Shrinkage processes that are also influenced by fibre orientation can lead to the compound becoming detached from the pin surface. In light of the 4 mm wall thickness of the sample, volume shrinkage is a factor that cannot be neglected. Pronounced shrinkage has to be expected particularly at points where the pin does not share the cavity cross-section.

As is clear from the SEM images in Fig. 10, macroscopic detachment results between the compound and the pin. In the case of samples produced with a high mold temperature, it is evident that the matrix material was not yet fully frozen at the time of detachment, which meant that contact with the surface was

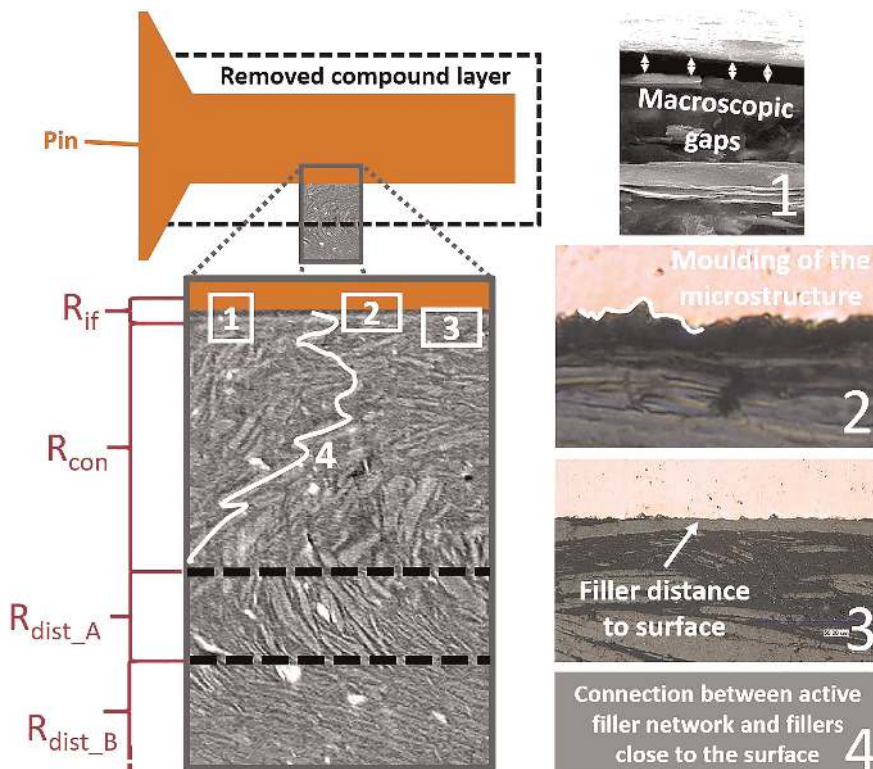


Fig. 9. Four key influences on the electrical contact resistance

maintained via individual filaments. As a general rule, detachment does not occur over the entire surface and remains in the range of just a few micrometres.

It is, however, clear that the detachment is not as pronounced with higher mold temperatures and hence also pin temperatures. Two factors are decisive for this: on the one hand, the slower cooling of the material means that the packing pressure has longer to act and overall shrinkage is less pronounced. On the other hand, the higher pin temperature leads to better mechanical interlocking of the matrix and the tin-plated surface (Fig. 11).

7.2 Microscopic Reproduction

The tin-plating process leads to a rough, porous surface on the pin as shown in Fig. 11 (left). It is thus not only possible for the surface to be reproduced but also for the matrix to penetrate the surface. Penetration of the porous surface by fillers is highly improbable on account of the small pore diameter (just a few micrometres).

At high mold temperatures, it was possible to see that the melt penetrates the rough surface structure (Fig. 11, right). This mechanical anchoring meant that samples produced with a higher mold temperature displayed cohesive fracture behavior to a greater extent when cryofractures were produced for microscopy (Fig. 12).

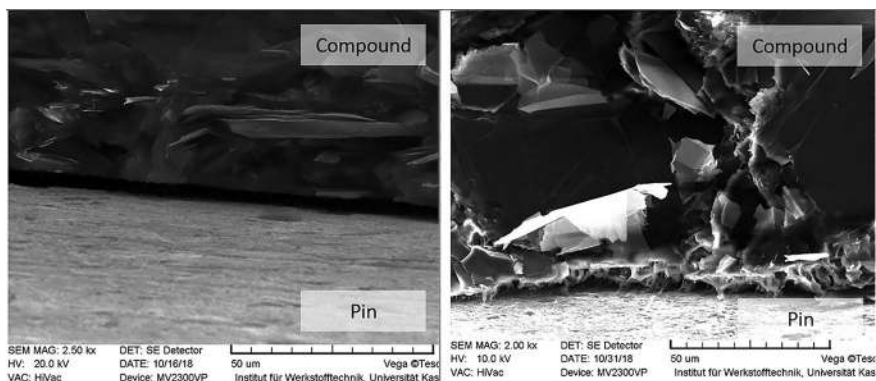


Fig. 10. Delamination between pin and compound, mold surface temperature (left) 40°C, PB3, and (right) 120°C, BP4

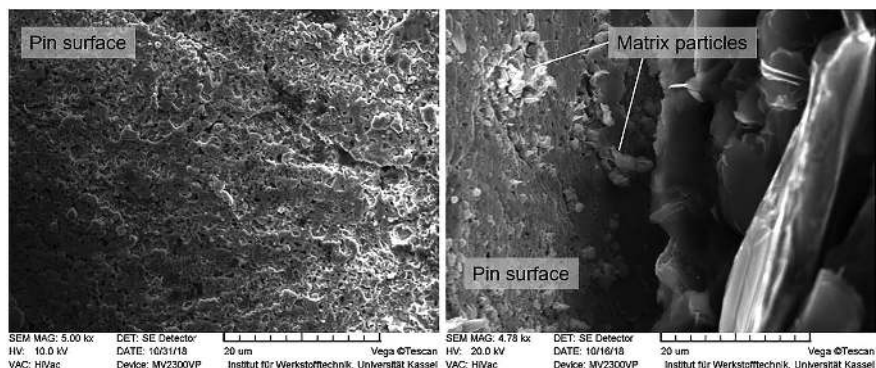


Fig. 11. Porous structure of the surface (left) and matrix particles in the surface (right)

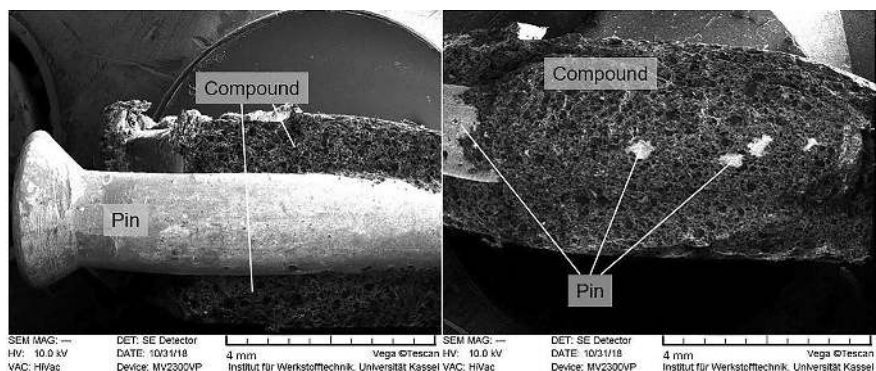


Fig. 12. Fracture behavior of samples produced with low (left) and high (right) mold temperature

7.3 Filler Position within the Interface

Two aspects must be borne in mind when observing the position of the fillers in the interface area. On the one hand, the minimal distance between a filler particle and the compound surface and hence the pin, and, on the other hand, the alignment of the particles in relation to the pin surface.

On all the samples produced, filler particles only penetrated the surface in isolated cases. An intact edge layer is thus present, with a low filler content. When examining the compound surface under the microscope on the samples fractured in the interface area to the pin, it was possible to see filler at the surface in some cases (Fig. 13).

The SEM images also indicate that the alignment of the fillers plays a role in whether the surface is penetrated or not. Fillers aligned orthogonally to the surface seem to penetrate the surface more frequently. As a rule, the graphite plaques run parallel to the surface, so that there is little electric conduction through the layers close to the surface. One exception to this is special flow situations, such as the forced weld line in the present case. Figure 14 shows the filling process, in diagram form, based on the flow lines and indicates the areas of the pins in which a weld line develops.

7.4 Connection of the Filler Particles Close to the Interfaces

In order to observe the fibre orientation, μ -CT images were made of those areas of the sample in which a weld line forms when the pin is suitably aligned. Orientation structures are highlighted in the upper half of Fig. 15. Inside the weld line, the fillers are aligned orthogonally to the pin surface, both close to the pin and also over the cross-section of the flow towards the

outside. Samples with pins aligned in the direction of flow thus display partially perpendicular fillers on the surface (Fig. 15, left). Samples with pins aligned counter to the direction of flow do not have any weld lines. Over the entire pin surface, the fillers are thus aligned parallel to this surface (Fig. 15, right).

As already discussed in the previous section, this changed orientation influences both the positioning of the particles directly in the interface and their connection to the rest of the network. In the region of the weld line, it is more probable that filler particles will penetrate the surface and, in addition, a better connection is achieved for the centre of the compound, since the electrons can be conducted into the centre of the material directly perpendicular to the interface along the fillers.

8 Conclusion

The investigations have shown that, even with materials that do not conduct so well, like the PP graphite compound presented here, the electrical contact resistance plays a key role in the overall electric behavior of an overmolded sample. For the samples observed, the line resistance would only attain the same order of magnitude as the sum of the two contact resistances with a pin spacing of approximately 200 mm.

The effects of weld lines for boosting electrical conductivity that are familiar from the literature can be successfully proven in the contact regions too. The positive effect is due, above all, to a better penetration of the interface and a better connection of the fillers that are closest to the edge to the overall network. The clearly lower contact resistances for samples produced with higher mold temperatures can be attributed to a better macroscopic and microscopic connection between the compound and the pin.

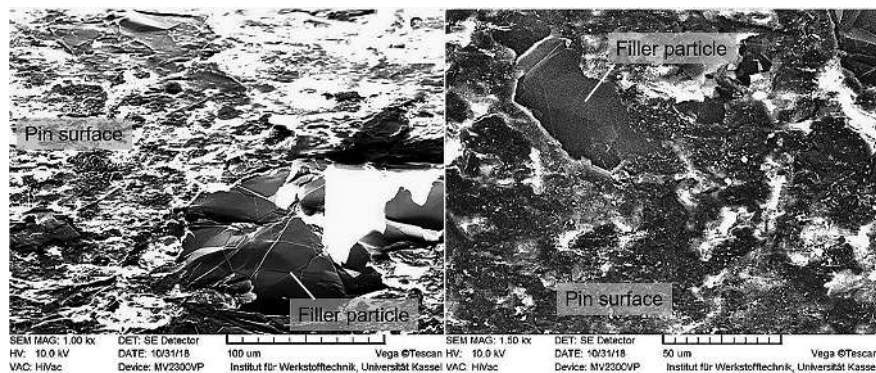


Fig. 13. Penetration of the surface by filler particles for samples produced with low (left) and high (right) mold temperatures

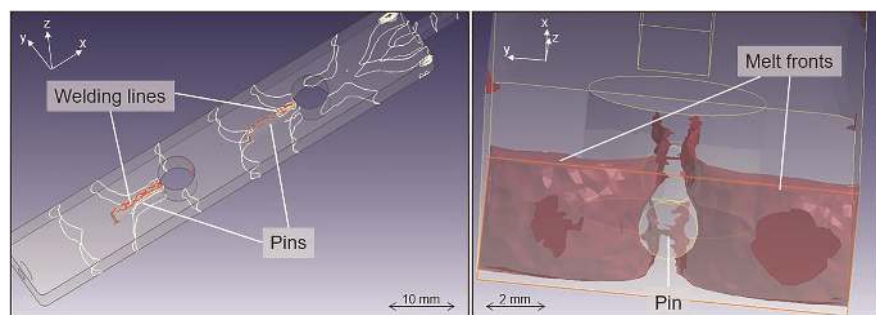


Fig. 14. Position of welding lines based on a flow simulation (left) and detailed view of the melt fronts prior to the weld line being formed above and below the pin

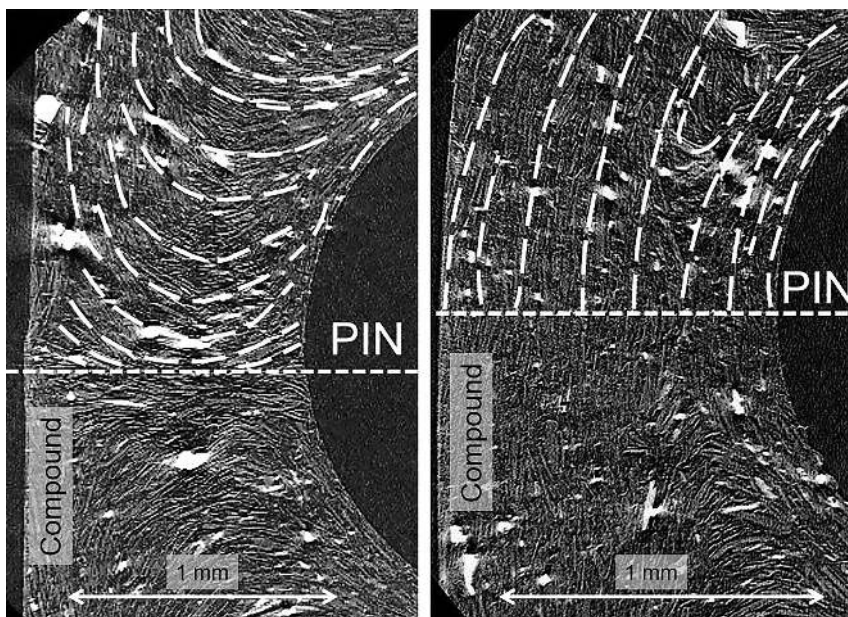


Fig. 15. Filler orientation depending on pin alignment in the flow direction (left) and against the flow direction (right)

The investigation was able to show the importance of electrical contact resistance in injection molding taking the example of a graphite filled polypropylene. Further investigations will have to observe the transposability of the results to other materials and fillers. Over the long term, this will enable design rules to be defined for injection molding contacts.

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