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# Innovative Materials for Novel Concepts for Internal Heat Exchangers

*Internal heat exchangers are often employed in solar thermal applications as well as in many other applications that involve heat, and, for the most part, consist of metallic materials (copper, stainless steel). Corrugated stainless steel tubes are easy to assemble, and are commonly used in various storage geometries. In order to significantly reduce costs, alternative materials, such as plastics, can function as excellent substitutes. Alternative heat exchangers for the following examinations using various materials and an extrusion process were produced. Subsequently, the heat exchangers were examined in regards to the heat they transfer. The heat transfer coefficient and the thermal conductivity of all materials were determined.*

## 1 Introduction

Internal heat exchangers are often employed in solar thermal applications as well as many other applications that involve heat, and, for the most part, consist of metallic materials (copper, stainless steel). Corrugated stainless steel tubes are easy to assemble, and are commonly used in various storage geometries. In order to significantly reduce costs, alternative materials, such as plastics, can function as excellent substitutes.

In general, plastics display very little capacity for conducting heat (0.15 to 0.5 W/(m K)). In contrast, metals display values ranging from 10 W/(m K) up to 400 W/(m K) (Amesöder and Ehrenstein, 2008). Incorporating conductive filling materials can lead to a significant improvement of the thermal conductivity. For example, if filling material grades of up to 60% are incorporated, the thermal conductivity can increase to values of up to 20 W/(m K) (Heinle and Ehrenstein, 2009; Kumlutas and Tavman, 2006). Thus far, metallic filling materials (i.e., copper), ceramic filling materials (i.e., boron nitride), and organic filling materials (i.e., graphite) have been favored.

The selection of the matrix material is an important aspect, since the thermal conductivity can vary strongly when using the same filling materials with differing matrix materials (Weidenfeller et al., 2002). In materials with a high filling content, the filling materials are densely packed, and, therefore, deter-

mine the property profile of the produced material (Öztürk, 2012). On the whole, the thermal conductivity in a compound depends on the level of contact amongst the individual molecules and particles. For this reason, it is important for thermal conductive particles to have according points of contact amongst one another (Posca, 2011; Hong et al., 2011). Figure 1 depicts an example illustration of a model of highly thermal conductive compounds with according, densely situated points of contact.

Filled plastics can achieve thermal conductivity levels between 1 and 20 W/(m K). These levels are more than sufficient for most technical applications. Highly thermal conductive plastics are subject to percolation, which means that the conductivity begins to increase once a specific filling material concentration has been achieved. Adding small amounts of filling materials does not have any effect and no noteworthy increase in the thermal conductivity results (Leute, 2015). Once the percolation threshold has been reached, the conductivity increases significantly.

Due to the small spaces between the materials, the incorporated filling materials can develop conductive network structures. However, percolation is dependent on numerous factors, which, in turn, makes it challenging to determine a specific threshold value for the according material combination. Depending on the type of filling material used and the amount, processing must be adapted accordingly, since the flow profile changes starkly. The mobility of the molecules decreases due

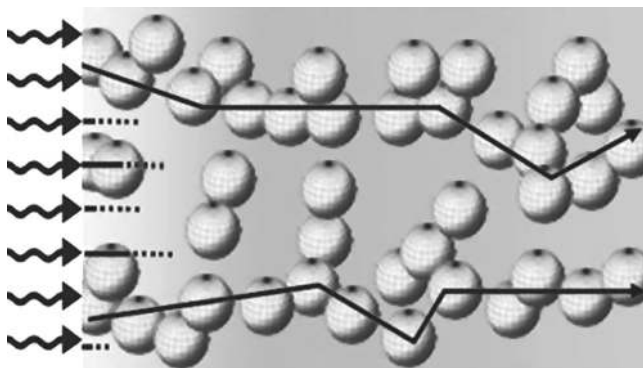


Fig. 1. Increase in the thermal conductivity owing to densely situated, conductive filling material particles (Posca, 2011, modified illustration)

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to the increased ratio of filling material, which results in a substantial increase in the viscosity (viscosity displacement factor) (Amesöder, 2009; Dörner, 2012).

Plastic heat exchangers have thus far been employed in various designs (plate heat exchangers, spiral tubing or tube bundles), but mainly are applied with corrosive media, i. e., in the chemical industry. The type of plastic that is selected correlates with the type of medium being used. These heat exchangers are very expensive owing to the fact that they almost always must be produced according to the specifications made by the customer. In contrast, thermally conductive composite materials that have rarely been employed up until now, display a high amount of technical and economic potential for heat exchanger applications.

Alternative heat exchangers for the following examinations using various materials and an extrusion process were produced. Subsequently, the heat exchangers were examined in regards to the heat they transfer. The heat transfer coefficient and the thermal conductivity of all materials were determined.

## 2 Alternative Heat Exchanger Concepts

Tubes made of plastic or even plastic composite tubes are used in many areas of application today. Examples include drinking water lines, heating tubes and floor or wall heaters made of polybutene (PB), PE-X (cross-linked PE), three-layer composite tube PE-RT-Alu-PE-RT, and PVC-C (PVC post-chlorinated).

Plastic heat exchangers with a greater wall thickness require materials with an accordingly higher level of thermal conductivity. If the wall thickness is thin, it is important to take into account that pressure stability problems can occur during assembly and handling.

In order to be able to assess which levels of thermal conductivity will be required for which wall thickness, the required heat exchanger length for defined boundary conditions was calculated.

The overall heat transfer coefficient can be calculated using the following Eq. 1 that consists of the three components of heat transfer (internal convective heat transfer, heat conduction and external convective heat transfer).

$$UA = \frac{2\pi L}{\frac{1}{\alpha_i \cdot r_i} + \frac{1}{\lambda} \ln\left(\frac{r_a}{r_i}\right) + \frac{1}{\alpha_a \cdot r_a}}, \quad (1)$$

with L: tube length,  $\alpha_i$ : convective heat transfer coefficient inside the tubes,  $r_i$ : internal radius of the tube,  $\lambda$ : thermal conductivity of the tube wall,  $r_a$ : external radius of the tube and  $\alpha_a$ : convective heat transfer coefficient outside the tubes.

The external convective heat transfer coefficient  $\alpha_a$  is determined using the Nusselt correlation (Eq. 2) for the external heat transfer in the case of free convection, which is triggered by a heated, horizontal tube in a water bath (VDI, 2006).

$$Nu = [0.752 + 0.387 \cdot (Gr \cdot Pr \cdot f_2)^{1/4}]^2. \quad (2)$$

The internal convective heat transfer coefficient  $\alpha_i$  describes the heat transfer from the fluid flowing in the tube to the inner wall of the tube. The Nusselt correlation recommended by

Gnielinski for turbulent tube flows was employed (Gnielinski, 1995).

$$Nu = \frac{\frac{f_1}{8} \cdot Re \cdot Pr}{1 + 12.7 \sqrt{\frac{f_1}{8}} \cdot (Pr^{1/4} - 1)}, \quad (3)$$

with the pressure loss coefficient  $f_1 = (1.8 \log_{10} Re - 1.5)^{-2}$ , L: tube length,  $d_i$ : inside diameter of the tube,  $Pr_w$ : Prandtl number at the tube wall. All material properties except for  $Pr_w$  are evaluated at the mean fluid temperature inside the tube.

Figure 2 provides the results of this calculation. The following boundary conditions were selected:

- 10 kW heat flux,
- 5 K temperature difference between the average fluid temperature in the heat exchanger and the average temperature in the storage element,
- External diameter of the tube: 25 mm.

The thermal conductivity capacity of the material in the calculation was varied from a low level to a high level of thermal conductivity (0.23 W/(m K) to 10 W/(mK)). The results are shown as a function of the wall thickness. The graph shows that the required heat exchanger length in the case of the selected boundary conditions is approximately 55 m at a high level of thermal conductivity (approx. 10 W/(m K)). The wall thickness has hardly any influence in this case. With case values of thermal conductivity the influence of the wall thickness becomes more significant, which requires larger lengths of the heat exchangers. A practically realistic maximum for the length is about 70 m. The wall thickness due to stiffness and strength as well as producibility ideally is in the range 300 – 600  $\mu\text{m}$ . These boundary conditions have been marked in Fig. 2. Thermal conductivity of approximately 1 W/(m K) would be sufficient for the named boundary conditions.

## 3 Experimental Details

In the following section, plastics that contain additives capable of conducting heat were selected. Three materials were pro-

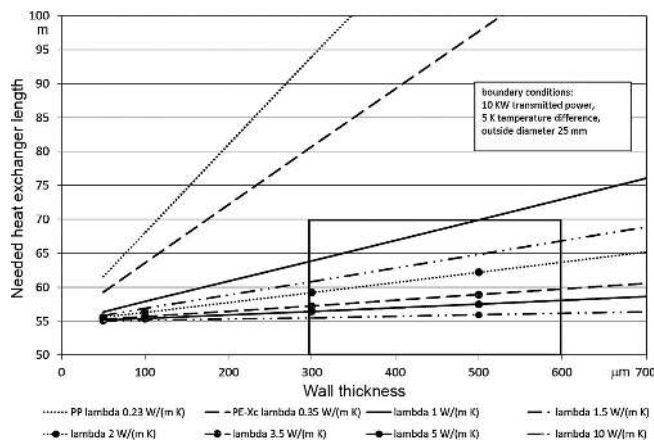


Fig. 2. Required length of thermal conductivity capacities and wall thickness (transferred power: 10 kW. Temperature difference: 5 K between the media in the heat exchanger and in the store, external diameter: 25 mm)

vided by two manufacturers, and these materials display a thermal conductivity that is higher than the theoretically determined 1 W/(m K) according to the data sheet. The used filled compounds are only suitable for the injection molding process. For better processing by extrusion, the filled compounds were mixed with a mixing ratio of 1 : 2 with matrix material and processed further. Thereafter the filler content of the produced tubes was determined by means of a thermogravimetric analysis according to DIN EN ISO 11385. Therefore all materials were heated to 900 °C. The remaining inert components represent the filler content. The filler content for all batches was 15 %.

Moreover the company Roth Werke GmbH, Dautphetal, Germany, provided another unfilled material, which is a stretched polyethylene (PE). The stretching in the material creates a special organization of the side chains, leading to a high level of flexibility in the material. This is meant to guarantee better handling of the hoses, even when the wall thicknesses are thinner.

The materials from Table 1 were processed into tubes in an extrusion process. Since commercially available compounds are mainly conceptualized for the injection molding process, a material with sufficient viscosity for extrusion was compounded.

The manufacture of the tubes was carried out by means of extrusion on a micro-extruder (self-construction, Dr.-Ing. I. Rbbelke, Geseke, Germany), which was equipped with an experimental device for hose and tube extrusion. In order to avoid damages caused by the material being pulled off the rollers, i. e., pinched-off seams, a vacuum-powered removal device was designed and constructed (Fig. 3) that allows the very thin-walled heat exchanger tubes to be pulled off with a defined external diameter (15 mm). In the following section, the thermal conductivity samples, which had wall thicknesses ranging between 0.3 and 0.5 mm, were examined.

#### 4 Examination of the Thermal Conductivity $\lambda$

In order to determine the thermal conductivity, the following individual parameters (with  $\lambda$ : thermal conductivity,  $a$ : tem-

perature conductivity,  $\rho$ : density and  $c_p$ : specific heat capacity) were determined in accordance with Eq. 4:

$$\lambda = a \times \rho \times c_p, \quad (4)$$

The determination of the specific heat capacity  $c_p$  was carried out three-fold using dynamic differential calorimetry (DSC analysis) with a DSC module, Q 1000, TA Instruments, Eschborn, Germany.

The determination of the density  $\rho$  was carried out using a buoyancy method with propanol ( $\rho = 0.787 \text{ g/cm}^3$ ) as a solution in a double determination. The calculation of the density was carried out in accordance with Eq. 5 (with  $\rho$ : density,  $m_L$ : weight in air,  $\rho_{FL}$ : density in propanol,  $m_{FL}$ : weight in liquid):

$$\rho = m_L \times \rho_{FL} / (m_L - m_{FL}). \quad (5)$$

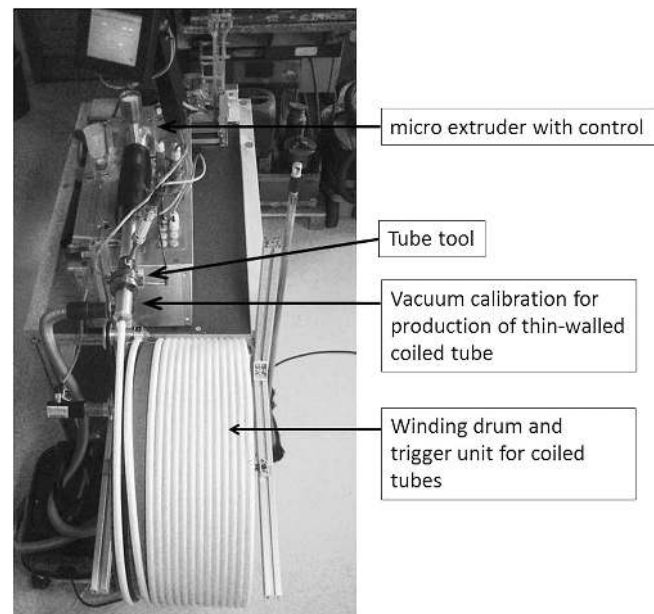


Fig. 3. Extrusion of filled polypropylene with a vacuum-powered removal device (plan view)

Material	Filling material/form of the filling material	Thermal conductivity W/(m K)	Filler content wt%
Lehmann & Voss & Co. LUVOCOM 105-8617 thermoplastic elastomer (TPE) unreinforced, modified for thermal conduction [graphite gray]	Graphite platelet form	9.5 (according to the data sheet)	12 (TGA analysis)
Lehmann & Voss & Co. LUVOCOM 65-8330 polypropylene (PP)-copolymer unreinforced, modified for thermal conduction [gray]	Graphite platelet form	3.4 (according to the data sheet)	15 (TGA analysis)
Albis Plastic GmbH polypropylene (PP) with Boron Nitride white	Boron nitride platelet form	3 (appr.-information provided by the manufacturer)	15 (TGA analysis)
Roth Werke GmbH PE-RT Type II Dowlex 2388 stretched polyethylene (PE) [white]	No thermal conductive filler	0.43 (according to the data sheet)	0 (TGA analysis)

Table 1. Employed materials, modified for thermal conductivity

The determination of the temperature conductivity was carried out with a light flash apparatus LFA 467 HyperFlash made by the company Netzsch, Ahlden, Germany. The prepared materials (50 mm × 50 mm) were sprayed with graphite prior to testing, and, subsequently, were measured. The temperature increase in the sample, which results from the absorption of a laser flash, is measured. The parallel bottom side of the sample that is to be measured is heated with an energy impulse. The resulting temperature change on the upper side of the sample is measured by means of an infrared detector. The measurements were carried out at room temperature. The device manufacturer gives an accuracy for the temperature conductivity of ±3%. Subsequently, the value is calculated directly by the software. Three laser flashes were measured and evaluated per sample.

The following measurements of the steady-state heat transfer of the heat exchanger samples were performed on a test station developed at the University of Kassel (Germany) with the support of the ISFH in Hameln, Germany (Fig. 4). The samples to be tested were 300 cm long and had varying wall thicknesses ranging from 250 μm to 1 300 μm.

Figure 5 provides a cross-sectional depiction of the development of a flow within the test station during the test of the sample in charging mode (heat input in the test station). The water is heated by the sample, and flows upward within the flow guide sheets. Once it reaches the top, it flows to both sides. While passing the cooling circuit, the water is cooled again. Once steady-state conditions have been reached, the sample is



Fig. 4. Test station for heat exchanger samples

hit with a flow from below at a constant temperature. The temperature is measured by 18 temperature sensors located above and below the test sample. Furthermore, the flow and return temperatures of the heating circuit are measured, as well as the volume flow through the sample. The accuracy of the volume flow rate measurements is < 0.5% of the measured value, the temperature sensors have an accuracy according to EN 60751 class B for flow and return temperatures and EN 60751 class A for the temperature sensors in the test tub. The following boundary conditions were applied for all measurements:

- Flow rate in the sample: 550 l/h,
- Flow temperature to the sample: 54 °C.

The required cooling temperature depends upon the heat exchanging capacity of the sample. Three different cooling temperatures were set per sample with the goal of achieving mean logarithmic temperature differences between 10 and 20 K.

### 5 Results and Discussion

The thermal conductivity capacities determined using Eq. 4 are listed in Table 2 for all measured materials. It is evident that the added conductive filling materials did not lead to a significant increase in the thermal conductivity in comparison to un-

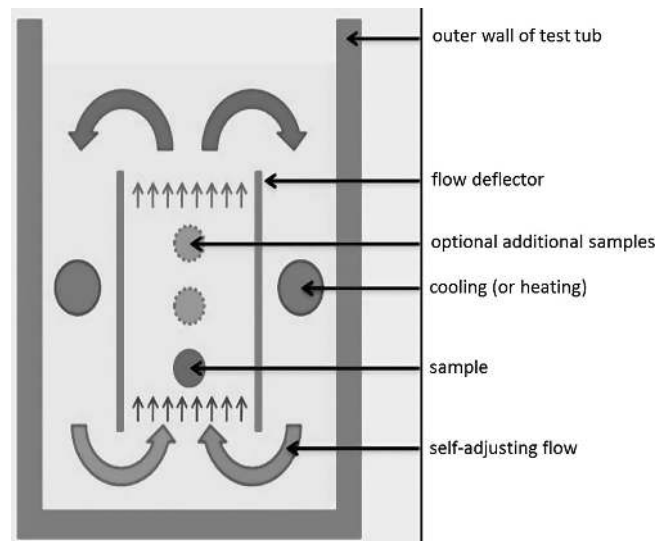


Fig. 5. Cross-section of the targeted flow in the water bath while operating in charging mode

Material	Thermal conductivity after the addition of a filling material W/m K	Thermal conductivity without a filling material W/m K
TPE + graphite	0.8	0.7
PP + graphite	0.2	0.2
PP + boron nitride	0.2	0.2
PE-RT (stretched, without a filling material)		0.4

Table 2. Thermal conductivity capacity results of all examined samples

filled materials. For one, this could be attributed to the material thickness. Also, it could be attributed to the reorientation of the filling materials during the extrusion process (here: platelet geometry, determined by means of ash accumulation and microscopy (Fig. 6)). The geometry of the filling materials is a significant factor for the thermal conductivity. It is higher on the platelet level (up to 400 W/(m K)) than perpendicular to that (2 W/(m K), in-plane, through-plane) (Skrabala and Bon-ten, 2015).

Figure 7 uses PE-RT as an example to show that the measured heat transfer coefficients increase as expected as the wall thickness reduces. In comparison to a copper tube, PE-RT with a wall thickness of approximately 0.25 mm achieves values that fall within the satisfactory range according to Fig. 2.

By means of the measured overall heat transfer coefficient UA and the calculated convective heat transfer coefficients  $\alpha_i$  and  $\alpha_a$ , the thermal conductivity  $\lambda$  can be calculated using Eqs. 1 to 3. A comparison of the measured values (Table 3) illustrates that the results derived from the heat transfer measurements are confirmed when compared with the values directly determined for the thermal conductivity from the density determination, the determination of the  $c_p$  value, and the temperature increase measurement.

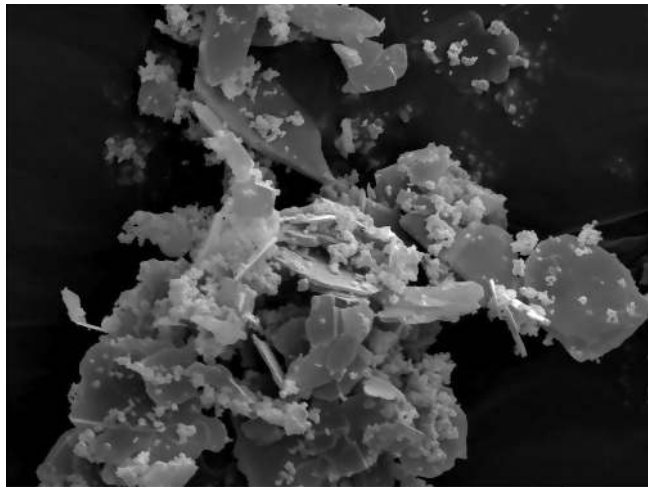
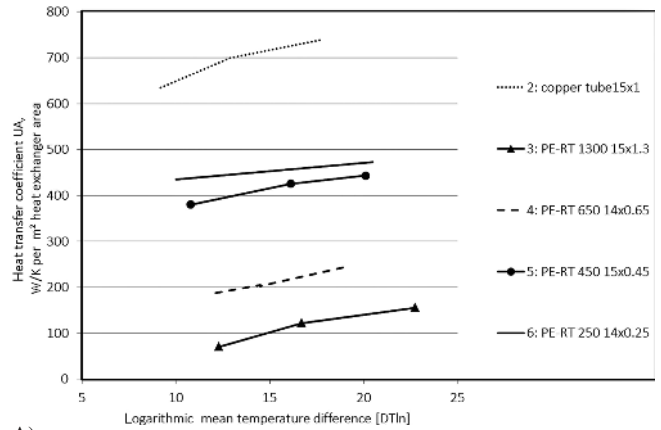
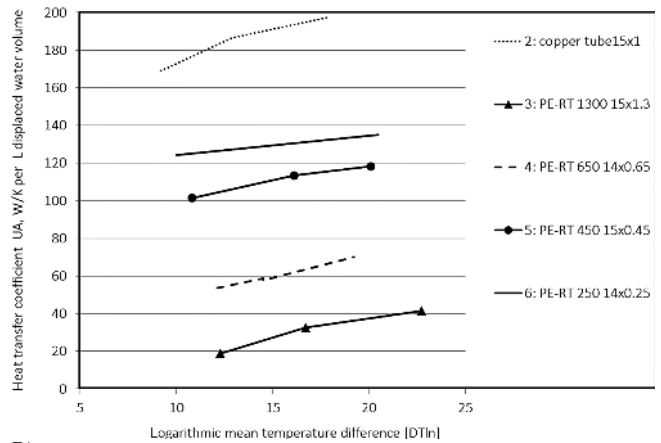


Fig. 6. Scanning electron microscopy: Determined platelet geometry of the PP sample with boron nitride, 1000 ×

Figure 7 shows curves for tubes with 15 mm diameter and for those with 14 mm diameter. In order to simplify comparison, the measurement results obtained for the 14 mm tubes were converted to an external diameter of 15 mm with the following calculation. Once the thermal conductivity levels of all measured hoses are known, Eqs. 1 to 3 can be used to convert the measurement values of the heat transfer coefficient to other hose diameters. This value was then used to adjust the measurement value accordingly, as can be seen in Fig. 8.



A)



B)

Fig. 7. Measured heat transfer coefficients of a copper tube sample measuring 15 × 1 mm and PE-RT tubes with different wall thicknesses, A) shown in W/K per m<sup>2</sup> of heat transfer area, B) shown in W/K and L displaced water volume)

Material	Thermal conductivity capacity determined based on the temperature conductivity W/m K	Thermal conductivity capacity determined based on the heat transition measurements W/m K
TPE [graphite gray]	0.8	0.9
PP gray	0.2	0.2
PP with boron nitride [white]	0.2	0.4
PE-RT	0.4	0.4

Table 3. Comparison of thermal conductivity capacities determined based on the temperature conductivity and heat transfer measurements

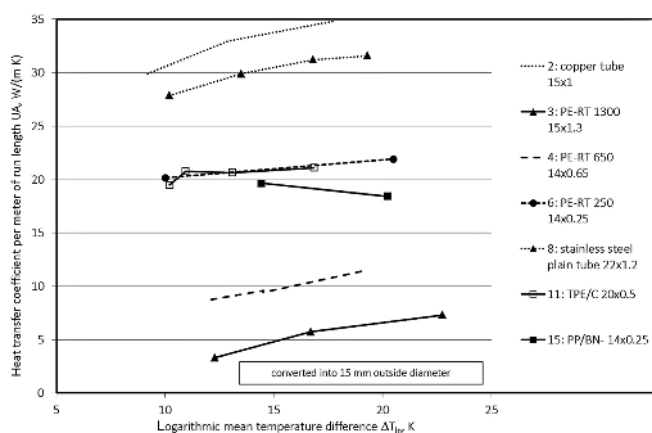


Fig. 8. Measurement results of the heat transfer coefficient per meter of the run length of various heat exchanger tubes for tubes with an external diameter of 15 mm. In part, tubes with other diameters were tested, and these values were theoretically converted to an external diameter of 15 mm

Figure 8 depicts the overall heat transfer coefficient in W/K and meter for its run length and shows the results of various heat exchanger tubes that were measured in the steady-state heat exchanger test station. The values of the copper and stainless steel tubes (as a function of the logarithmic mean temperature difference between the medium in the tube and the medium in the test tub) are approximately 30 W/(m K). If thin walls measuring 250  $\mu\text{m}$  were present, as in PE-RT as well as PP with boron nitride, values were determined that lay within the range of 20 W/(m K). As expected, thicker wall thicknesses (650 or 1300  $\mu\text{m}$  in PE-RT) resulted in significantly lower heat transfer coefficients. Interestingly and in contrast, there is the result of TPE/C with a wall thickness of 500  $\mu\text{m}$ . In this case, the thermal conductivity is considerably higher than in an unfilled plastic, and lies within the range of very thin-walled, unfilled hoses.

## 6 Conclusions

The manufactured plastic tubes display a very good heat transfer coefficient when they have very thin wall thicknesses ranging from 0.2 to 0.3 mm. Such tubes are relatively difficult to work with in real applications due to the risk of leakage. Several difficulties when producing highly thermally conductive, filled plastics were encountered. First, the plastic types must be suited for extrusion in regards to their viscosity. Plastics available from compounders are usually only intended for use in injection molding processes. For this reason, the material manufacturers provided small amounts of compounds with a higher viscosity that was suitable for extrusion.

Furthermore, additives are often incorporated in platelet form. They orient themselves along the tube in the direction of the flow during the extrusion process. The thermal conductivity along the tube is good, but not in the perpendicular direction, which is required in a heat exchanger. Also, the so-called percolation threshold has to be surpassed. This means that the thermal conductivity only begins to increase significantly once

a certain filling ratio has been reached which allows the filling materials to develop conductive network structures owing to the short distance. In the case of platelets this threshold is on a different level for in-plane and through-plane measurements and strongly depends on the extrusion conditions. Depending on the type and content of the used filler the technological process has to be adapted according to the material flow behavior. The mobility of the molecules decreases due to an increased filler content. This can lead to a significant viscosity factor (viscosity displacement factor). Materials which have flowability suitable for the process should be used. In order to keep the filler content as low as possible with sufficient thermal conductivity, the conception of a tool with a modified melt flow for extrusion, which could lead to a reorientation of the fillers, would be useful.

Several promising measurement results were able to be obtained in the performed tests. By employing TPE as the matrix material, a high level of bendability was able to be achieved despite the added filling materials. Nonetheless, processing was relatively complicated. In order to improve the mechanical properties (strength, pressure stability), a good matrix-filler connection is required. In this case, coupling agents can provide a remedy. Coextrusion of multilayer hoses could also lead to a more effective functional bond and thus to an increase in mechanical properties.

Surprisingly, the simple, unfilled PE-RT tube, as is commonly used in floor heating, was another satisfactory variant. The wall thickness would have to be maximally 0.65 mm in order to guarantee utilizable heat transfer (compare to Fig. 7). The advantages in this case are the exceptionally low material and manufacturing costs.

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