CHARACTERISTICS OF NOVEL POLYMER COMPOSITE HEAT TRANSFER TUBES

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ABSTRACT

Heat transfer tubes are one of the basic components in many equipment widely used in chemical, pharmaceutical, petroleum, petrochemicals, power generation and air conditioning systems. These tubes are manufactured from a wide variety of high thermally conductive metals. Examples of such materials are: nickel alloys, copper, aluminium, stainless steels like austenitic, duplex and super duplex as well as super alloys and titanium. Metallic tubes may suffer from failure due to corrosion and erosion especially in aggressive environments. In addition, metallic tubes are characterized by high density and high costs. This is the motivation for the development of non-metallic materials for heat exchanger applications.

Plastics are attractive materials for the construction of heat exchangers, especially in harsh environments. However, plastics have one essential negative character; low thermal conductivity.

There are two different approaches to overcome these drawbacks. On the one hand the use of small tube/channel diameter and extraordinary small wall thickness can reduce the thermal resistance. On the other hand the more promising solution is to improve the thermal conductivity by filling plastics with very high thermally conductive materials.

This paper provides an overview of different plastic materials suitable for use in heat transfer applications. Also, it summarizes the properties of different possible filling materials. The thermal conductivity of these composite materials strongly depends on the size, shape, mass fraction and orientation of the particles. The possibilities to control the particle orientation of filling materials within the wall of a plastic tube will be shown. The effect of filling material orientation on thermal conductivity of tubes in axial and orthogonal direction and the influence of the wall thickness are illustrated.

The paper compares the properties of developed tubes with customary used heat transfer tubes. Properties compared in this paper include fouling and corrosion resistance, thermal and mechanical properties, erosion resistance and temperature limitations. The novel polymer composite heat exchanger tubes combine a high thermal conductivity, low density, excellent resistance against ionic corrosion with an outstanding resistance against abrasion and erosion.

INTRODUCTION

Heat exchangers are widely used in many industries such as power generation, air conditioning, petroleum, petrochemicals and pharmaceutical. The basic elements in this equipment are heat transfer tubes, which enable the heat exchange between two or more fluids at different temperatures. Currently these tubes are manufactured from a wide variety of high thermally conductive metals. Examples are nickel alloys, copper, aluminium, austenitic stainless steels, duplex, super duplex, super alloys and titanium. Main drawbacks of these metallic tubes are high density, low chemical resistance, high rate of erosion and fluctuation in cost. On the other hand, tubes made of plastics or composite polymers are inert, resist corrosion and erosion, are light in weight and can be shaped easily. However, they have other unfavourable properties for heat transfer applications. Heat conductivity is a disruptive factor for the practice of polymers in heat exchanger tubes. Operation temperature of standard plastics is relatively low for most of engineering applications.

On the other hand, there are two different approaches to overcome the problem of low thermal conductivity of plastic materials. The first one is the use of very thin wall heat transfer surfaces. The second tactic is the use of composite polymer materials, which are polymer filled with extraordinary high thermally conductive additives. The filling material has to be inert to heat transfer fluids in order to evade negative effects like corrosion, pollution and scaling.

NOMENCLATURE

k	[W/mK]	Thermal conductivity
x	[-]	Content of fillers
U	[W/m²K]	Thermal transmission coefficient
h	$[10^{-6}*1/K]$	Heat transfer coefficient
t	[mm]	Thickness (wall)
R_f	[m²K/W]	Fouling resistance

Special characters		Subscripts	
		P	Polymer
A	Surface area	C	Compound
T	Temperature	i	inner
d	Diameter	o	outer
Q	Heat	T	Tube

BACKGROUND

The first extensive research in the field of plastic heat exchangers started in the 1960s [1]. Unfilled fluorine-polymers were used for tubes. They had a high resistance against chemicals, especially acids, but the heat conductivity of the material was really low. To increase the thermal flow of unfilled plastics the wall thickness was reduced to a minimum and the surface of the heat exchanger was enhanced. Subsequently, plastic heat exchangers have been used for heat recovery systems in polluted and corrosive fluids [2, 3]. Later for sea water desalination [4, 5], fuel cells [6], solar water heating [7] and other heat exchanger applications in the industry. In 1999 the influence of anisotropy was shown in the example of Nylon 6.6 and graphite [8]. During the early 2000s the first heat conductive modifications of thermoplastic polymers were tested [9, 10]. These materials were used for heat exchangers in gas-liquid heat transfer applications.

The thermal conductivity of polymer composite materials depends on many variables. This includes the type of polymer material, nature of the filling particles and the sizes of the filling elements. Similarly, it depends on the relation between polymer and fillers, the allocation of the filler in the polymer, the quantity of air pockets, the orientation of the filling elements and the aspect ratio (the relation between the mean diameter and the thickness of the particle for the anisotropic fillers).

POLYMERS FOR HEAT TRANSFER

Polymers are large organic molecules consisting of a series of repeating subunits. These units are called monomers and are connected to each other in the form of a chain. Polymer nomenclature is generally based upon the type of monomer comprising the polymer. Caused by the different monomers and crystallinity structure polymers establish a broad range of properties. Synthetic polymers may consist of crystalline and amorphous regions. The properties of polymers are characterized by their degree of crystallinity, ranging from completely amorphous to a theoretically crystalline polymer. Polymers with crystalline areas are generally tougher and show more impact resistance than very amorphous polymers. Amorphous polymers tend to be transparent or at least translucent.

The polymer material for heat exchanger applications has to fit to the combination of thermal, chemical, mechanical and custom-designed properties of the application.

The most important property data of plastics for the use in heat exchanger applications include thermal conductivity, specific heat capacity, and maximum allowable operating temperature, coefficient of thermal expansion, tensile strength, tensile modulus and density. Furthermore, thermoplastics show creep behaviour over time. For many polymer materials, long time data on lifetime and creeping are not readily available, indicating a need for further research to ensure the effectiveness of these materials.

One of the basic properties for the use in heat exchanger applications is the service temperature. The continuous operating temperature of the polymer has to be higher than the service temperature of the heat exchanger.

Caused by their chain structure, their binding energy and their texture there are polymers with higher and lower continuous operating temperatures available. Additionally heat stabilizers, modifier and other additives can change this value. Table 1 shows the thermal limitations for a long time usage of different polymers.

Table 1 thermal properties of different polymers

	melting temperature [°C]	thermal conductivity [W/mK]	continuous operating temperature [°C]
PP-H	163	0,22	60 (80)
PK	220	0,23	80 (140)
PPS	305	0,3	200 (210)
PVDF	175	0,19	140
PFA	205	0,22	240

A typical classification of plastics is shown in figure 1. Compounds made of commodity plastics like polypropylene (PP) can be used for applications up to 60 °C and under special conditions up to 80 °C. The optimal engineering plastic for heat exchanger applications up to 80 °C is polyketone (PK). Polyketone compounds can be used under certain conditions up to 120 °C. The best high performance plastic for heat exchanger applications is polyphenylensulfide (PPS). Polyphenylensulfide based compounds can be used up to 200 °C.

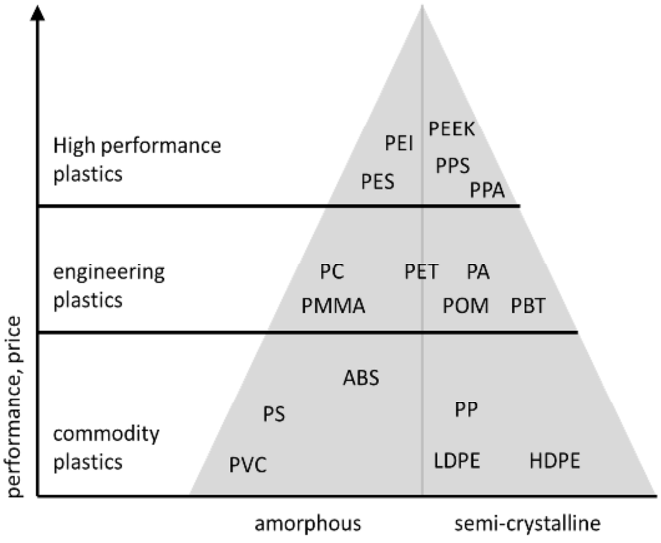

Figure 1 pyramid of thermoplastic polymers

In addition, fluoropolymers like polyvinylidene fluoride (PVDF) and perfluoroalkoxy alkanes (PFA) are conceivable for a use in heat exchangers.

A special compound solution can be realised by the use of high end polymers like polyetheretherketone (PEEK). The thermal resistance of PEEK goes up to 250 °C, but this material has an extraordinary price. Therefore it is a special solution for very high requirements only. Polyamide (PA) compounds can be used up to 120 °C, but polyamides cannot resist acid conditions, which are often used for cleaning or are part of the processing fluids. Also, Polyesters cannot be used - they show hydrolysis with hot water and Polycarbonates (PC) can create micro cracks in hot water environments. By the use of polymers, chemicals like anti-fouling agents, cleaning agents and anti-scaling agents can be reduced or avoided. If these additives can be avoided also other polymer materials can be the best choice for heat exchanger applications as well.

Another basic property for polymeric heat exchangers is the chemical resistance. Various applications and packages are made of plastics, because of their inert behaviour. The polymer structure and the formulation of polymer chains generate chemical resistance. Some materials like Polypropylene (PP) resist acids; other like Polyamides (PA) will be dissolved in acid environments. The polymer system has to be well selected for the particular environmental condition.

FILLERS FOR THE COMPOUND

In general, nearly all plastics can be filled with thermal conductive fillers. Excluded are polymers that do not allow high content of fillers in the extrusion process like LCPs, polymers that show a reaction with a filling material and the environment or polymers whose chains will be destroyed by the fillers during the processing.

Many types of filler are available in a spherical or ball-shaped form. However, this form reduces the generation of conductive paths of fillers in the surrounding material. The heat conductivity here is independent of the orientation and show isotropic behaviour.

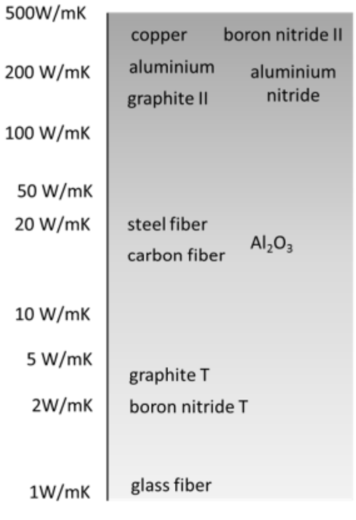

Figure 2 Typical thermal conductivity values of different fillers

The most effective and approved fillers show the disadvantage of anisotropic properties. Caused by standard production processes, anisotropic behaviour of such fillers prevents the increase of heat conductivity in a vertical direction to the wall of heat transfer tubes. The influence of the novel, engineered production process can massively prevent this effect. Thereby a cost effective, technical competitive product to metal tubes is generated.

Typical properties of fillers are shown in Figure 2. Some materials like graphite or boron nitride show high thermal conductivities in their longitudinal direction (II), but relatively poor values in cross direction (T). The values differ in two decades. If these materials are compounded together with plastic materials, an anisotropic material can be created. These are the best materials to increase the thermal conductivity by extrusion, if processing allows an orientation of the fillers.

FEATURES OF THE FILLING MATERIALS

The form, orientation and size of filling particles have strong impact on the thermal conductivity of the composite materials. For example, silicon carbide (SiC) has nearly the same thermal conductivity like boron nitride (BN), approximately 300 W/mK, but the thermal conductivity of composite polymer using this material is low, because of their smaller particle sizes.

The shapes of the most thermal conductive fillers are irregular and the particles differ in sizes as shown in figure 3.

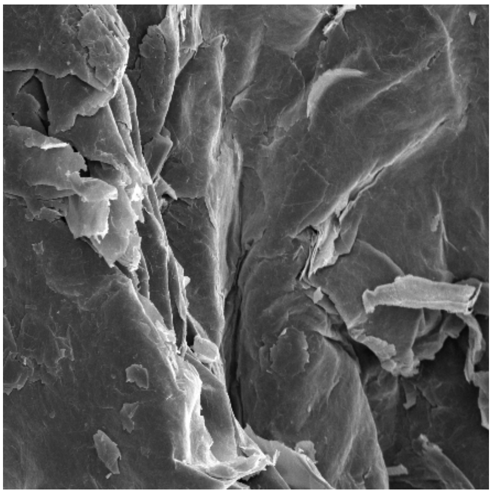

Figure 3 Scanning Electronic Microscope (SEM) pictures of an unshaped particle.

The unshaped surface of a thermal conductive particle is shown. The thermal energy in heat conductive plastic flows along the areas of highest heat conductivity; these are the filler particles in the thermal compound. Therefore, the orientation of the fillers is a major influence parameter on the controlling of the thermal conductivity of the composite. Also, the way of the thermal flow is shown in figure 4. The polymer in the compound acts as an insulator. For highest heat conductivity, the conductive particles are oriented in the heat flow direction.

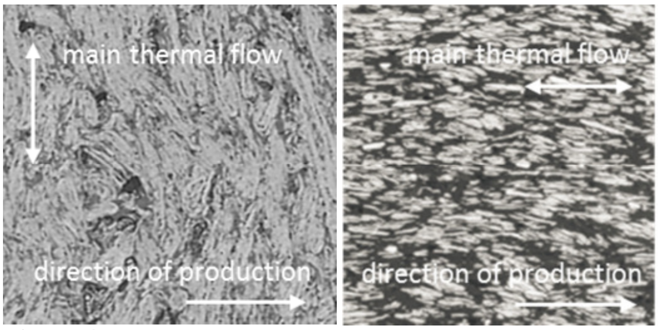

Figure 4 main thermal flow in a conductive compound
Left: Extrusion with particle-orientation, **right:** extrusion without particle-orientation

During a standard extrusion process of plastics and composites the most filling particles are oriented in extrusion direction. This improves heat transfer in longitude direction of the tube. However, heat transfer in the direction normal to the pipe axis will be low. By using an extrusion process that influences the particle orientation, a tube with orthogonal oriented particles can be generated. But not a completely orthogonal oriented product is the best in the thermal performance.

Conductive particles have to create a network of conductive fillers within the plastic. An orientation of 60 % of particles over 45 ° towards the extrusion direction can increase the thermal conductivity of a plastic graphite compound by 150 % [5]. This network of conductive particles is responsible for this effect. It is necessary for a high thermal conductivity.

EFFECT OF TUBE THICKNESS

A main factor to improve the rate of heat transfer is to minimize the thickness of the wall. The thickness is limited by the lowest mechanical requirements. Scheffler and Leao made investigations to test the heat transfer of thin, unfilled plastic films [11]. Films of 20 µm and lower showed similar performance to 1 mm titan tubes or CuNi-Alloys. To fulfill mechanical requirements in heat exchanger units a thicker wall plastic tube has to be used. This plastic tube has to be thermally modified to reach thermal performances of metallic tubes.

Figure 5 shows the effect of wall thickness and thermal conductivity on the overall heat transfer coefficient. The figure shows that the thickness has a high influence on heat transfer rates. On the other side, thermal conductivity of the novel composite depends additionally on the thermal conductivity of filling materials, the amount and orientation of filling materials and the number of voids.

To modify the orientation of fillers in order to improve thermal conductivity, a minimum wall thickness is required. The interaction between geometry of the fillers, viscosity of polymers, process parameters and the melt flow situation in the tool is very complex. All known mathematical models, are too imprecise to calculate this system or need quite hard border conditions. Therefore the heat conductivity of a heat conductive polymer compound has to be measured in experiments using real samples.

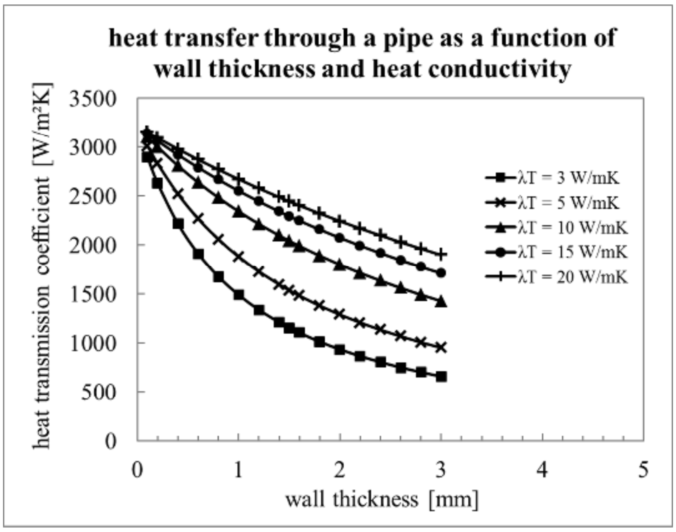

Figure 5 Heat transfer through a pipe as a function of wall thickness and heat conductivity

In 2009 Chen, Li and Guo [12] have shown that the thermal conductivity of a plastic tube system of about 15 – 20 W/mK can achieve 95 % of the performance of a titanium based heat exchanger system as shown in Table 2.

Table 2 Performance comparison between heat exchanger made of different materials, Chen at al. [12]

heat exchanger material	k [W/mK]	U_0	$U_o/U_{o,Ti}$	$U_o/U_{o,Al}$	$U_o/U_{o,Cu}$
modified PP	15	33,56	0,95	0,846	0,841
	16	33,89	0,959	0,854	0,849
	17	34,19	0,967	0,861	0,857
	18	34,46	0,975	0,868	0,864
titanium (Ti)	22	35,34	1	0,89	0,886
Aluminium (Al)	236	39,69	1,12	1	0,995
Copper (Cu)	398	39,9	1,13	1,01	1

For systems with low convective heat transfer coefficients such as air/air or air/vapor heat exchangers heat conductive composite tubes values are close to the values of metallic tubes. A thermal conductivity of above 10 W/mK shows a nearly comparable performance to titanium, aluminium and copper under these conditions. In systems with higher values of individual coefficient like liquid/liquid heat exchangers, heat conductivity of plastics cannot achieve the thermal performance of a metal based heat exchanger.

Table 3 properties of heat exchanger materials

material	density [g/cm³]	specific heat capacity [J/gK]	Thermal elongation [10^{-6} 1/K]	Flexual modulus [GPa]	thermal conductivity [W/mK]
Polypropylen - graphite	1,56	1,21	29	10,4	4-25
Aluminium	2,7	0,9	23,1	70	160-235
stainless steel	7,86	0,48	10-17	210	15-20
cupper	8,8-8,92	0,38	16,5	120	400
titanium	4,5	0,52	8,6	105	22
alubrass	8-8,35	0,37	18,7	90	112

Furthermore, plastic based heat exchanger elements can have a positive effect on the heat transfer coefficients due to their surface energy or the wettability of elements. For example the forcing droplet condensation with much higher heat transfer coefficient in a vapor condensing system.

Some major properties of different heat exchanger materials compares to the compound are shown in table 3.

TECHNOFORM HEAT CONDUCTIVE TUBES

Technoform Kunststoffprofile GmbH (TKP) located in Lohfelden, Germany develops and produces complex extrusion profiles for technical practice in the industry. Challenging geometries, calibrated hollow chambers, demanding precision and surface finish, as well as processing of extraordinary plastics are their fields of expertise. This company can produce high quality composite tubes made of thermoplastic materials and graphite with filler content of about 70 % per mass. Theses tubes are produced by a new developed, high-end thermoplastic extrusion processes. In this process, the orientation of fillers can be modified.

Extrusion of polymers, in contrast to extrusion of metals, is a continuous process lasting as long as raw material is supplied. Compounds can be used in the extrusion process like other plastic materials, too. An advanced process control is necessary to avoid damage of particles in highly filled compound materials. Additionally, special equipment is needed to generate a selective, continuous orientation of the filling particles to improve the thermal conductivity.

The overall heat transfer coefficient of thermal conductive polymer composite tubes is measured in an evaporator test rig. The influence of wall thickness and particle orientation was analysed. To compare the influence of the particles orientation on the thermal conductivity, different wall thicknesses were used. The metal tubes, used for a comparison, show, based on their metal structure, a constant thermal conductivity for tubes with different wall thicknesses. Tubes of different thickness made of polypropylene and graphite differ in their thermal conductivity values. This is caused by the production process, the influence on the fibre orientation and the distribution of the fillers in the material.

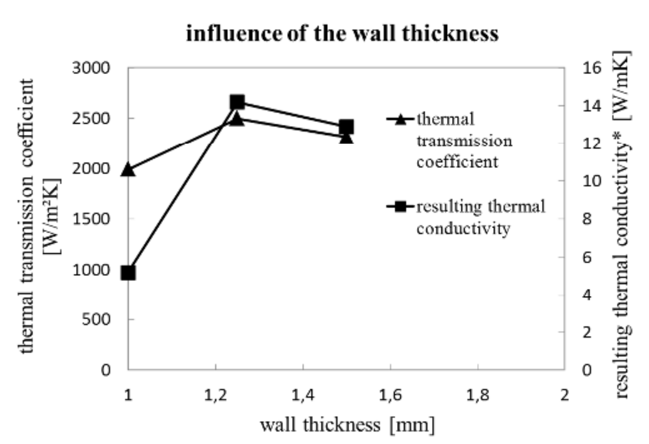

Figure 6 influence of the wall thickness of a heat conductive plastic on the thermal conductivity

In Figure 6 it is shown that the thermal conductivity of 1 mm tubes is worse than the thermal conductivity of 1,25 mm and 1,5 mm tubes. The thermal transmission coefficient should be much higher, based on the lower thermal resistance of thinner tubes, but the influence of the particle orientation dominates here.

It can be assumed that an efficient orientation of the heat conductive particles in the direction of the thermal flow is not possible for a tube wall thickness of 1 mm. The results from figure 6 were confirmed by investigations of microscope pictures of the tubes.

The particles can only be orientated in an orthogonal direction to the tubes, if following conditions are fulfilled:

- Optimized polymer-particle dimension relationship
- Sufficient ratio between the size of particles and the wall thickness
- Low viscosity of melt to enable an orientation of the particles
- Sufficient shear strain in the melt to induce an orientation.

COMPARISON OF METAL AND PLASTIC

The thermal performance of the plastic tubes was compared to metal ones in a long-time test under brine conditions. Scaling and fouling of the brine at the surface of tubes show a strong influence on the test results.

Table 4 thermal transition coefficient of different materials after long time test in brine environment

Heat exchanger material	wall thickness [mm]	transmission coefficient [W/m²K]
Polypropylen with graphite	1,5	2316
Polypropylen with graphite	1,25	2500
Polypropylen with graphite	1	1990
stainless steel	0,5	2289
titanium	0,5	2883

Although, the wall thickness of the composite tubes is 2,5-times thicker than the wall thickness of the metal tubes, the thermal transmission coefficient of 1,25 mm plastic tubes is higher than the stainless steel tubes and at least 87% of titanium tubes.

The density of the metal tubes is up to 5-times higher than the density of the polymer composite. Although, a slightly bigger surface area may be needed for plastic heat exchangers, but they still offer a better price due to their lower density.

CORROSION RESISTANCE

Corrosion is the gradual destruction of materials by chemical reaction with their environment. Corrosion process of metals is caused by electro-chemical reactions with an oxidant. This type of damage typically produces oxides or salts of the original metal. Corrosion in this form cannot arise in polymers.

Polymers can have degradation as well, but this is caused by other effects like e.g. ultraviolet radiation. Therefore plastics can be used under conditions where metals corrode. Metals are ranked according to their potential showed in figure 7.

The diagram shows materials with their electrical potential referenced to the Standard Calomel Electrode (SCE). Anodic materials like magnesium and aluminum tend more to be attacked than cathodic materials like titanium and graphite.

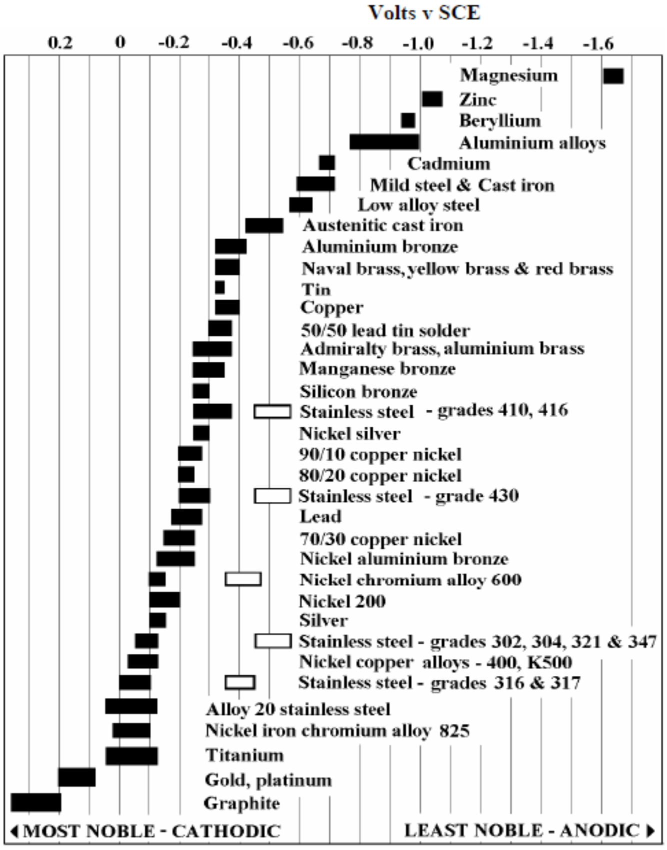

Figure 7 corrosion potentials in flowing sea water at ambient temperature [13]

The critical point for a galvanic corrosion is the contact of materials with difference in potential in an electrically conductive fluid environment. Already a difference of above 200 mV can create galvanic corrosion in an electrolyte environment. Some metals like zinc, magnesium, and cadmium show naturally slow reaction kinetics, even though their corrosion is thermodynamically favorable. While corrosion of these metals is continuous and ongoing, it happens at an acceptably slow rate. An extreme example is graphite. It has such slow kinetics that it is effectively immune to electrochemical corrosion under normal conditions. In figure 8 the surface modification of different materials after 100 days storage in industrial brine is shown.

Figure 8 stainless steel, PP-graphite and alubrass (left to right) after 100 days storage in industrial brine

It is well known and reviewed, that metals tend to corrosion under special conditions. Therefore plastic covered metallic tubes were analyzed to avoid corrosion of metallic tubes in brine, acid or hydrous environments. Polymer sheets between 19 – 25 µm are used for covering. The results show that thermal conductivity was reduced by 75 % [14]. Polymer coverings of metal tubes avoid corrosion but decrease the thermal conductivity massively. Additionally, there is always danger of delamination.

EROSION AND SCALE FORMATION RESISTANCE

The resistance against erosion was tested in the Faculty of Engineering in the Al-Imam Mohammad Ibn Saud Islamic University. In comparison to metallic tubes plastics tubes show a significant better resistance. The tubes were tested at very high velocities ranging from 2.5 m/s to about 20 m/s. The tests used the Riyadh city domestic water, synthetic sea water and sea water with different salt concentrations. The data obtained showed that the composite graphite tube can resist erosion to a high extent.

Plastic graphite compounds massively avoid scaling in seawater desalination plants. The deposition of salts on Polypropylene-graphite (PP-GR) and different metals was tested in the experimental station at the University of Bremen, Germany. In figure 9 the amount of deposition of Ca^{2+} and Mg^{2+} is shown.

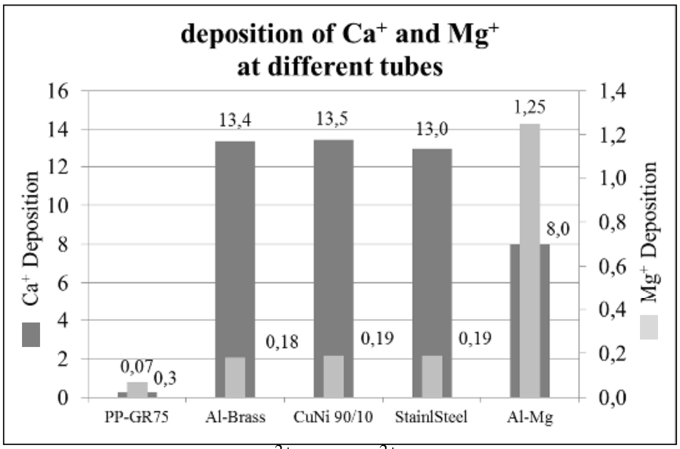

Figure 9 deposition of Ca^{2+} and Mg^{2+} at different tubes

The amount of deposits on the surface of PP-GR tubes is significant smaller than the amount of deposits on metal tubes. The test was carried out with artificial seawater (composition according to Kester et al. with salinity of 65 g/kg), 50h and a temperature of 75°C. These parameters are equal to the situation on the lower tubes in MED-plants for sea water desalination.

The results show a much better fouling resistance with use of polymer tubes. The low content of deposition on the polymer tubes can be removed easily by wiping. There was no crystallisation fouling which formed with the metals. A reduced scaling leads to lower costs due to fewer shut-downs for cleaning processes caused by the scaling as well as the advantage of using less anti-scalants.

Heat exchangers are used for many applications, in all areas of applications the total fouling related costs for major industrialized nation is estimated to exceed 4,4 Billion US-$ per year [15]. With the use of heat conductive polymer compounds this could be reduced dramatically.

Environmental protection also limits the use of copper in river, lake and ocean waters, since copper is toxic to aquatic life [16]. The actual daily discharge from desalination plants may amount to 18 kg of copper into the Mediterranean Sea, 74 kg of copper into the Red Sea and 296 kg of copper into the Gulf from MSF desalination plants only [17].

CONCLUSIONS

It is not recommended to use pure polymer materials for heat exchanger tubes because of performance reasons. Composite polymers filled with high thermally conductive particles can have a thermal conductivity close to that of titanium and stainless steel. The thermal characteristics of composite materials depend on the particle size, distribution, mass fraction, type and the orientation of filling particles.

Modern plastic process technologies allow the production of high thermally conductive plastic heat exchanger elements such as tubes or other shapes. The novel polymer composite heat exchanger tubes, developed from Technoform Kunstoffprofile GmbH, combine a high thermal conductivity, low density, excellent resistance against ionic corrosion with an outstanding resistance against abrasion and erosion.

Additionally, fouling and scaling in sea- and wastewater environment can be massively reduced and the use of chemicals for cleaning and fouling prevention can be avoided.

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