Engineered composite polymer sheets with enhanced thermal conductivity

Oguzhan Der¹, Stuart Edwardson², Marco Marengo³ and Volfango Bertola

¹Laboratory of Technical Physics, School of Engineering, University of Liverpool, Liverpool L69 3GH, United Kingdom
²Centre for Laser Manufacturing, School of Engineering, University of Liverpool, Liverpool LG9 3GH, United Kingdom
³School of Computing, Engineering and Mathematics, University of Brighton, Brighton BN2 4GJ, United Kingdom

E-mail: Volfango.Bertola@liverpool.ac.uk

Abstract. An innovative type of engineered composite polymer sheet with enhanced thermal conductivity is described and tested. The concept is to sandwich a closed-loop serpentine channel, cut out in a polypropylene sheet and containing a self-propelled liquid-vapour mixture, between two sheets of the same material bonded by selective laser welding. Polymer materials are widely used to replace metals in different applications, however sometimes this is not possible because of their poor thermal conductivity. Thin, flexible and low-weight polymer sheets can be advantageous in various aerospace, aircraft and portable electronic applications where the device weight and its mechanical flexibility are crucial. The transient and steady-state thermal response of a prototype engineered polymer sheet was characterised for different heat power levels and spatial orientations. The equivalent thermal conductance, calculated from the surface temperatures at opposite ends of the sheet, increases four to six times in comparison with a composite polypropylene sheet without working fluid.

1. Introduction

The use of polymeric materials to replace metallic parts is the obvious choice to address weight and cost constraints in a large number of devices and applications, including space, aircraft and portable electronics applications. Whilst polymeric materials offer excellent features of mechanical flexibility, resistance to fatigue, low weight and low cost in comparison with metallic materials, they exhibit poor heat transfer performances due to their low thermal conductivity, which for commercial polymers is often less than 1-2 W/mK.

Recently, there were several attempts to increase the thermal conductivity of polymers by means of high-thermal conductivity additives and fillers, such as minerals, fibres and metals [1]. The fillers size often reaches the nanometric scale to obtain a direct and efficient interaction between the composite components. The final properties of such nanocomposites depend on the properties of their components and on the shape, size, distribution and orientation of the dispersed phase. Fillers can be added in-situ during the synthesis process or ex-situ by mechanically dispersing the filler into a molten polymer using a turbomixer or a twin-screw extruder. Commonly used fillers include particles [2, 3], fibres [4], metal powders or particles [5, 6], and carbon nanotubes [7-9].
In the present work, it is proposed to enhance the thermal conductivity of polymer sheets by embedding a self-driven liquid-vapour mixture, which transfers heat from an evaporator to a condenser region of the material according to the well-known working principle of pulsating (or oscillating) heat pipes (PHP) [10]. The pulsating heat pipe is a thermally driven heat transfer device, which usually consists of a long tube in a serpentine configuration, which is firstly evacuated and then partially filled with a working fluid. Due to capillarity, the working fluid initially resides as an alternation of liquid plugs and vapour slugs. When heat is applied to the evaporator zone, the fluid inside the tube is activated: liquid plugs receive sensible heat while vapour slugs expand due to evaporation and push the heated fluid towards the condenser zone. There, vapour slugs undergo condensation, releasing heat to a cold source. This promotes a pulsating/circulating slug flow.

A simple and cost-effective manufacturing process to embed the heat transfer fluid into composite polymer sheets is described. Using this process, a number of prototype composite polypropylene sheets were fabricated and tested. Results indicate the equivalent thermal conductivity of the engineered composite material shows a four- to six-fold increase with respect to the conductivity of the composite polypropylene sheet without working fluid.

2. Design and manufacturing
Prototype composite sheets consisted of one black polypropylene layer (0.7 mm thick) sandwiched between two transparent polypropylene layers (0.4 mm thick), all with a length of 250 mm and a width of 98 mm. The material was chosen based on: (i) mechanical properties (elastic modules, yield stress, resistance to fatigue); (ii) compatibility with a range of organic heat transfer fluids (ethanol, acetone, refrigerant fluids); (iii) suitability to laser cutting and welding process. The maximum continuous service temperature is 130°C.

A serpentine shaped channel was cut-out in the central black layer, and featured either three or five turns, as illustrated in Figure 1. The channel had a width of 9 mm, which was determined so that the hydraulic diameter, \( D_H = 1.3 \text{ mm} \), satisfies the design criterion given in Eq. (1), which ensures surface forces prevail on gravity [11]:

\[
0.7 \frac{\sigma}{g(\rho_L - \rho_G)} \leq D_H \leq 1.8 \frac{\sigma}{g(\rho_L - \rho_G)}
\]

Figure 1. Technical drawing of the five-turns channel (a) and top view of the three-turns channel cut-out in the polypropylene sheet (b).

Figure 2. Schematic of the PHP assembly and laser welding process.
According to Eq. (1) the hydraulic diameter depends on the fluid properties; in particular, for ethanol ($\rho = 789$ kg/m$^3$; $\sigma = 22$ mN/m) Eq. (1) yields $1.18 \text{ mm} \leq D_h \leq 3.04 \text{ mm}$, while for the refrigerant fluid FC-72 ($\rho = 1680$ kg/m$^3$; $\sigma = 10$ mN/m) the criterion becomes $0.54 \text{ mm} \leq D_h \leq 1.4 \text{ mm}$. The five-turns channel (Figure 1a) had a total volume of 8.4 ml, while the three-turns channel (Figure 1b) had a total volume of 5 ml.

The transparent polypropylene layers were bonded on the two sides of the channel by selective laser welding [12, 13], as shown schematically in Figure 2, to create a strong and seamless joint between adjacent layers; while the external sheets are transparent to the laser wavelength, the central black layer absorbs the same wavelength, to enable polymer bonding exactly at the interface, without affecting the rest of the material. Two polypropylene rings were glued (Loctite All Plastic) in correspondence of two holes in one of the transparent sheets, to fit a pressure transducer and a micro-metering valve used for introducing the heat transfer fluids (FC-72 for the five-turns channel and ethanol for the three-turns channel, respectively).

3. Experimental setup and procedure

A schematic view of the experimental setup is displayed in Figure 3. The polymer sheet sample was mounted on a vertical support, with the heated zone (evaporator) at the bottom and the cooled zone (condenser) at the top. The heat supply was provided to both sides of the evaporator by two ceramic heaters (100 W each); two copper plates were used to distribute heat uniformly, and a thin layer of heat sink paste was applied on surfaces to minimise the contact resistance. The heaters were connected to a regulated DC power supply (Circuit Specialists CSI 120001X). Two fan-assisted heat sinks (Malico) were used to remove heat from the condenser section.

The polymer sheet was connected to a pressure transducer (GEMS 3500, 0-160kPa) and to a micro-metering valve (Upchurch Scientific); the pressure transducer DC output was sampled at 1 Hz by a data acquisition system (LabJack U6). Eight surface K-thermocouples (Omega Engineering) were placed on the heat pipe surface, four in the evaporator and four in the condenser zone. The temperature distribution in the adiabatic region was monitored by a FLIR One infra-red camera (FLIR Systems Inc.).

To introduce the working fluids (ethanol and refrigerant FC-72), composite sheets were vacuumed to a pressure of 0.2±0.5 kPa (abs) using a two-stage vacuum pump (Bacoeng) connected to the micro-metering valve; after disconnecting the pump, the valve was connected to an external reservoir.
containing the heat transfer fluid and gently re-opened until the desired amount (3 ml for ethanol and 4 ml for FC-72, corresponding to 60% and 50% of the total hollow volumes, respectively) flowed into the composite sheet.

Experiments were conducted by applying to the evaporator section an ascending/descending stepped heating power ramp ranging approximately between 2 W and 30 W, and measuring the temperatures on the composite polymer sheet surface. For each power step, the heat supply was kept constant until a pseudo steady-state regime was attained. Tests were interrupted earlier in case any point of the material reached a temperature of 110°C.

The equivalent thermal conductance of the engineered composite polypropylene sheet in the longitudinal direction was calculated as:

\[ K_{eq} = \frac{\dot{Q}}{T_{ev} - T_{cond}} \]  

where \( T_{ev} \) and \( T_{cond} \) are the averages of the four thermocouples temperatures measurements of the evaporator and condenser sections, respectively, and \( \dot{Q} \) is the heating power supply.

4. Results

An example of the temperature distribution in the adiabatic region when the composite polymer sheet is heated in the evaporator zone, obtained from FLIR measurements, is shown in Figure 5. Unlike materials that are homogeneous in the direction of the heat flux, where one can identify a temperature gradient with isothermal lines orthogonal to the heat flux, the engineered composite sheet displays alternating hot and cold stripes, corresponding to the ascending boiling fluid and the descending condensate, respectively.

Temperatures measured in the evaporator and in the condenser zones of the composite polymer sheet during the ascending/descending heating power supply ramp are displayed in Figure 6. Due to the relatively small thermal conductivity of polypropylene, and to the intrinsically unstable gas-vapour flow, the composite sheet exhibits significant thermal inertia, and reaches a pseudo-steady state in about 40 minutes after each step change in the heating power supply.

![Figure 5. FLIR images of the adiabatic region during the heating ramp.](image)

![Figure 6. Temperatures measured in the evaporator and in the condenser zones of the composite sheet during the ascending/descending power supply ramp.](image)
The overall thermal performance of the composite polymer sheet is shown in Figures 7 and 8, which display the equivalent thermal conductance of the material between a hot end and a cold end, calculated according to Eq. (2), as a function of the heating power supply for the three-turn-channel sheet filled with ethanol and the five-turn-channel sheet filled with FC-72, respectively. To quantify the heat transfer enhancement due to the fluid circulation, these values are compared with the equivalent thermal conductance of the vacuumed composite sheets without heat transfer fluid.

The highest value of the equivalent thermal conductance is observed for the five-turn-channel sheet filled with FC-72, at the maximum heating power supply of 31.3 W, where it attains a maximum value of 0.74 W/°C; this means the equivalent thermal conductance of the PHS increases of 585% with respect to the composite polypropylene sheet without working fluid (0.126 W/°C). The corresponding increase of thermal conductivity observed in the three-turn-channel sheet filled with ethanol is about 250% higher in comparison with the composite sheet without working fluid (0.067 W/°C), i.e. 0.16 W/°C at the maximum heating power supply of 14.7 W. The different performance is obviously due to the different number of turns of the channel, and the different fluid properties, which make evaporation of FC-72 (heat capacity: 1.1 kJ/kgK; latent heat; 88 kJ/kg) easier compared with ethanol (heat capacity: 2.5 kJ/kgK; latent heat; 846 kJ/kg).

In both cases, the maximum performance is limited by the temperature of the composite sheet in the evaporator region, which is limited by the maximum continuous service temperature of the material used.

5. Conclusions
A novel concept of engineered composite polymer sheet was designed and manufactured using three polypropylene sheets, bonded together by selective laser welding, where the central sheet contains a serpentine channel filled with a heat transfer fluid. The thermal response was evaluated for two different geometries of the channel and two different fluids, for different values of the heat input at the evaporator.

Preliminary results indicate a 250% increase of the equivalent thermal conductance for the three-turn-channel sheet filled with ethanol and a 585% increase for the five-turn-channel sheet filled with FC-72, respectively. The proposed technology represents a promising route to produce composite polymeric materials with enhanced thermal characteristics.

![Figure 7](image1.png)  
**Figure 7.** Equivalent thermal conductance of three-turn sheet filled with ethanol, as a function of the heat rate supplied at the evaporator. The dashed line indicates the thermal conductance of the composite sheet without fluid.

![Figure 8](image2.png)  
**Figure 8.** Equivalent thermal conductance of five-turn sheet filled with FC-72, as a function of the heat rate supplied at the evaporator. The dashed line indicates the thermal conductance of the composite sheet without fluid.
Acknowledgments
O. Der gratefully acknowledges a YLSY doctoral studentship from the Republic of Turkey, Ministry of National Education.

References
[1] Mazumadar S K 2002 Composite manufacturing: materials, product and process engineering Boca Raton, Fla, CRC Press London https://www.taylorfrancis.com/books/9781420041989
[2] Kumluetas D and Tavman I H 2006 Journal of Thermoplastic Composite Materials 19 (4) 441 https://doi.org/10.1177/0892705706062203
[3] Krupa I, Boudenne A and Ibos L 2007 European Polymer Journal 43 (6) 2443 https://doi.org/10.1016/j.eurpolymj.2007.03.032
[4] Tavman I H, Akinci H 2000 International Communications in Heat and Mass Transfer 27 (2) 253 https://doi.org/10.1016/S0735-1933(00)00106-8
[5] Maiti S N, Ghosh K 1994 Journal of Applied Polymer Science 52 (8) 1091 https://doi.org/10.1002/app.1994.070520810
[6] Chae D W, Hwang S S, Hong S M, Hong S P, Cho B G, Kim B C 2007 Influence of High Contents of Silver Nanoparticles on the Physical Properties of Poly(Vinylidene Fluoride) Taylor & Francis Great Britain 464 233 https://tandfonline.com/doi/abs/10.1080/15421400601031140
[7] Biercuk M J, Llaguno M C, Radosavljevic M, Hyun J K, Johnson A T, Fischer J E 2002 Applied Physics Letters 80 2767 https://doi.org/10.1063/1.1469696
[8] Hagedenmueller R, Guthy C, Lukes J R, Fischer J E, Winey K I 2007 Macromolecules 40 (7) 2417 https://pubs.acs.org/doi/abs/10.1021/ma0615046
[9] Winey K I, Kashiwagi T, Mu M 2007 MRS Materials Research Society 32 348 https://doi.org/10.1557/mrs2007.234
[10] Ma H 2015 Oscillating Heat Piper Springer New York https://link.springer.com/book/10.1007/978-1-4939-2504-9
[11] Paudel S B, Michna G J 2014 ASME Proceedings ICNMM2014-22016 V001T05A006 https://doi.org/10.1115/ICNMM2014-22016
[12] Acherjee B Kuar A, Mitra S, Misra D 2015 International Journal of Advanced Manufacturing Technology 78 (5-8) 853 http://doi.org/10.1007/s00170-014-6693-7
[13] Lui H, Jiang H, Chen G, Guo D, Yan Z, Li P, Wang X 2016 International Journal of Advanced Manufacturing Technology 86 (1-4) 809 http://doi.org/10.1007/s00170-015-8242-4