University of Plymouth

PEARL **https://pearl.plymouth.ac.uk**

Faculty of Science and Engineering School of Engineering, Computing and Mathematics

2023-03-10

3D heated mould tool development for the manufacture of PLA matrix composites via in situ polymerization (ISP) during monomer infusion under flexible tooling (MIFT)

Qin, Y

https://pearl.plymouth.ac.uk/handle/10026.1/20839

All content in PEARL is protected by copyright law. Author manuscripts are made available in accordance with publisher policies. Please cite only the published version using the details provided on the item record or document. In the absence of an open licence (e.g. Creative Commons), permissions for further reuse of content should be sought from the publisher or author.

Project acronym: SeaBioComp

Project title: Development and demonstrators of
durable biobased composites for a marine environment.

Project Number 2S06-006

D 1.4.2

3D heated mould tool development for the manufacture of PLA matrix composites via in situ polymerization (ISP) during monomer infusion under flexible tooling (MIFT)

Due delivery date:

Actual final delivery date: 10 March 2023

Organization name of lead participant for this deliverable:

University ofPlymouth

Dissemination level		
PU	Public	
CO	Confidential , only for members of the consortium	X

SeaBioComp is an Interreg 2 Seas 2014-2020 project
covering England, France, the Netherlands and Belgium.

Deliverable number:	D 1.4.2
Deliverable responsible:	University of Plymouth
Work package:	WP1

Author(s) – in alphabetical order (principal authors in bold)		
Name	Organisation	E-mail
Cullen, Richard	University of Plymouth	R.Cullen@plymouth.ac.uk
Graham-Jones, Jasper	University of Plymouth	jasper.graham-jones@plymouth.ac.uk
Meng, Maozhou	University of Plymouth	maozhou.meng@plymouth.ac.uk
Pemberton, Richard	University of Plymouth	richard.pemberton@plymouth.ac.uk
Qin, Yang	University of Plymouth	yang.qin@plymouth.ac.uk
Summerscales, John	University of Plymouth	J.Summerscales@plymouth.ac.uk

Document Revision History			
Version	Date	Modifications Introduced	
		Modification Reason	Modified by
v1	29 July 2021	First draft	YQ and JS - UoPlymouth
v2	10 March 2023	Final version	JS - UoPlymouth

3D heated mould tool development for the manufacture of PLA matrix composites via in situ polymerization (ISP) during monomer infusion under flexible tooling (MIFT)

Yang Qin and John Summerscales

School of Engineering, Computing and Mathematics (SECaM), University of Plymouth, Plymouth PL4 8AA, United Kingdom.

Abstract

Due to the greater awareness of the environmental issues, the use of "greener" materials to substitute for synthetic fibre reinforcements and petrochemical polymer matrices is growing rapidly. Therefore, to minimise the ecological impact, the SeaBioComp project aims to produce natural fibre bio-based thermoplastic composites for marine applications. The University of Plymouth is developing monomer infusion under flexible tooling (MIFT) for the manufacture of bio-based composites via in situ polymerisation (ISP). The monomer material selection identified methyl methacrylate (MMA) for PMMA and L-lactide for PLA as potential matrix systems for marine composites. MMA can be processed at ambient temperature in a similar manner to conventional thermoset epoxy and polyester resins. However, L-lactide requires an elevated temperature between 100–200°C. There is a trend in using out of oven/autoclave processing for composite production due to the advantages including energy saving, lower equipment and operational costs, flexible in temperature adjustment, etc. Therefore, integrally heated mould tool attracts increasing attention, especially for large composite structure production.

This report reviews the suitable materials for the mould tool and heating elements available on the market. Most importantly, the design of a 3D heated mould tool for an offshore wind turbine nacelle cover demonstrator is presented, and the achievement of sensible thermal uniformity has been investigated using an infrared camera as the temperature has a significant influence on the property of the final composite structure.

Keywords: Thermoplastic; Monomer, Infusion; Heated Mould Tool.

1 Introduction

As an economical, clean method, Resin Infusion under Flexible Tooling (RIFT) has been widely used for the manufacture of large composite structures with high fibre volume fraction. RIFT is similar to Resin Transfer Moulding (RTM), with one mould face replaced by a flexible vacuum film [1-3]. In RIFT, it is common to flood one surface of the laminate with resin in a flow medium (a.k.a. transport mesh) to expedite fill by through-thickness flow [4]. In this SeaBioComp project, the University of Plymouth is using the RIFT method to produce natural fibre reinforced thermoplastic matrix marine composites via in situ polymerisation (ISP). The process will be referred to as Monomer Infusion under Flexible Tooling (MIFT). By considering the requirement of viscosity, water uptake, processing temperature, etc., methyl methacrylate (MMA) and L-lactide were reported to be suitable monomer candidates for ISP [5]. Similar to conventional thermoset resins, the process for MMA is straightforward and can be conducted at ambient temperature. It is anticipated that bio-based MMA will be commercially available in the near future, it is not available as a bio-monomer yet in this project. L-lactide monomer is produced from the fermentation of 100% natural and renewable agricultural resources [6], nevertheless high processing temperature (usually 100–200 °C) is required for the MIFT process. Furthermore, the ISP temperature has a significant influence on the molecular weight, conversion rate, crystallinity of the final polylactic acid (PLA) polymer, which in consequence will affect the properties of PLA matrix composite [7]. Therefore, maintenance of the uniformity of thermal field will be a key factor in composite production.

One of the major objectives by University of Plymouth in SeaBioComp project is to produce a 3D natural fibre reinforced thermoplastic composite demonstrator by MIFT via ISP. To enable composite production based on both MMA & L-lactide monomers, and also provide experiences for the out of oven/autoclave, a 3D heated mould tool with sensible temperature uniformity is discussed and developed in this report.

Copyright © SeaBioComp

2 Mould tools

2.1 Mould tool materials

A good mould tool is crucial for the successful, effective, and efficient manufacture of a 3D composite structure. As discussed in D1.4.1.2, "The tool should have adequate rigidity to maintain dimensional tolerances, generous curvature at corners to avoid fibre bridging, and often needs to be vacuum-tight for specific processes." The normal materials utilised for mould tools for composite production include: metal, wood, composites etc. [8].

Stewart [9] reported a trend towards lighter-weight, durable composite tooling due to the increase industrial demand of large-scale composite. Invar, a nickel-steel alloy, has long been the tooling material of choice for high-volume runs of composite parts because of its durability and a coefficient of thermal expansion close to that of the reinforced plastics. However, with the increasing size of the composite structure, the disadvantage of excessive weight and high cost in machining the metal made composite engineers more interested in using carbon fibre composite as the tooling material. Stewart also summarised that bismaleimide (BMI), epoxy and novel low-shrink resin systems can all be used for the composite tooling, with the highest working temperature up to 250°C, as well as the advantages including repairable, dimensionally stable and the ability to hold vacuum integrity. DURATOOL 450 BMI/carbon fibre tooling prepreg from Cytec Engineered Materials [10], HTM 556 and HTM 515-1 tooling prepregs from the Advanced Composites Group (ACG) [11] and HX series of low temperature epoxy tooling prepregs provided by Amber Composites [12] are all available composite prepregs for composite tooling on the market. There are also commercial available machinable core materials for composite tooling, for example: GrafTech International's GRAFOAM® carbon foam [13], CB1100 ceramic tooling block material from ACG [11], a structural material CFOAM® made from coal provided by Touchstone Research Laboratory [14], etc.

2.2 Heating systems

It was reported in deliverable D.1.4.1.2 "Heated mould tool development for bio-based composite manufacture" that the heating techniques include:

- conductive heat transfer using (gas-, oil or steam-) heated (metal) bodies
- convection of heated (inert) gas
- combined radiation and convection heating using an open flame
- local or scanned laser radiation
- ceramic long-to-medium wavelength infrared heaters
- quartz medium-to-short wavelength lamps
- microwave, induction or radio-frequency heating
- electrical cartridge or resistance elements
- ultrasonic heating

Follow the discussion in D.1.4.1.4, this report will also focus on the out of oven conductive heating using embedded heating elements. Out of oven heating method usually possesses the advantages of energy saving and more rapid heating, as it minimises parasitic heating of air in the oven. Therefore, the production method is more suitable towards "green" composites. In addition, out of oven is more suitable for the production of large composites production, e.g. wind turbine blade, as it has no limitation of the structural dimension and saves the capital investment of the expensive equipment.

Heated fabrics are most widely used for out-of-oven integrally heated mould tools. The heating elements can be either positioned at the primary tool face or flexible secondary tool face in MIFT process.

The commercial available heated fabrics include:

1. Fibretemp® system [15] is an energy-efficient temperature control system for moulds in fibre reinforced composite manufacturing. By applying an electric potential on electroconductive carbon fibres (with much lower conductivity than metallic materials such as aluminium or copper), they can be used as a heat source in resistance heaters. Heat is generated through the electric resistance of the carbon fibres. The very low thermal expansion coefficient of carbon fibres ensures a high dimensional stability of the mould in a corresponding structure, so that they remain free of distortion and have exceptional dimensional stability in the mould even at high temperature

Copyright © SeaBioComp

gradients. Due to the good drapability of heating carbon fibre fabrics, complex geometries can be equipped with Fibretemp® system, application temperatures up to 260 °C have already been achieved.

2. Tibgrid® [16] heating fabric provided by TIBTECH uses PTFE, MFA, PFA or equivalent coated heating wires. This heating system possesses surface power ranging from 500 to 7000 Watts/m², and is capable of achieving a maximum temperature of 240°C. Other applications of Tibgrid® include aircraft de-icing and pre-heating of composite fuel tank, fluid transfer pipes, engines or battery casing in cold regions. Furthermore, TIBTECH also supplies another elastic heating textile Tibgrid-Stretch, which is made by attaching the electrical heating circuit to an elastic fabric. It allows easier layout on complex shapes, or to follow curves on a flat surface. Therefore, it can be perfectly adapted to the complex shape composite manufacture out-of-autoclave.

3. CoTexx® [17] knitted heating fabric is an electrical resistance heating made of copper litz (enamelled) wire. The knitting of fine resistance wires makes CoTexx® flexible, therefore adaptable to complex contours. This product is able to achieve a maximum working temperature of 200°C with the benefit of identical heating power at any location and no hot spots. By means of CoTexx®, self-heated-mould for the production of composite structures can be manufactured or integrated. The benefit of this product according to the supplier includes: (1) no hot spot due to parallel connection, (2) stable regulating circuit for the temperature control, (3) gentle transfer of large heating capacities, etc. CoTexx® can be used for (1) heating of tools for FRP manufacturing, (2) de-icing in aerospace applications.

Heated fluids

Another heating method is heated fluid circulation through channels or piping mounted on the tool back surface. This is a well-established heating technique by utilising the circulating water or oil through the pipes (usually embedded on the back of the tool) [18,19]. Hot oil circulation heating of metal tooling can achieve high heating rates and has been applied in the wind energy field. Another major advantage of this fluid heating system is that can be used reversely, to cool the tool/composite following curing. However, special hoses and tubes are necessary, this will increase the costs for the heating system significantly; and the pipeline work is also time and labour consuming. In addition, the difference in temperature between the outlet and inlet pipe is also a problem [20].

The Quickstep process developed by Quickstep Technologies (Australia) should be mentioned here [21,22]. Instead of the air in the oven, this method utilises a liquid heat transfer fluid to heat or cool the composite structure. In the Quickstep process, the tool with the uncured composite component are sealed in a vacuum bag and then placed in a low-pressure pressure chamber of the heating/cooling liquid. This method has the benefits of rapid heating/cooling and lower tooling cost in composite production; nevertheless, Quickstep is still a liquid in-oven method in principle as there is still a limitation in composite dimension and a large proportion of the energy is consumed in heating/cooling the liquid fluid. In consequence, the Quickstep method will not be applied in this report.

Other heating technology

Buckypaper is a thin sheet (porous structure) made from an aggregate of carbon nanotubes, and its composites could be applied in the electrically conductive fields [23,24]. Tarfaoui et al [25] developed self-heating/deicing composites using Buckypaper within the composite. It was observed that the composite can be heated up in a short time-period by an electrical power, which implied an excellent heating performance of the Buckypaper. It was also speculated that Buckypaper may exhibit a better thermal conducting performance not only in the plane but also more importantly along the thickness direction, compared with graphite films [26]. Nguyen et al [27] have successfully prepared carbon fibre/Buckypaper hybrid composites by in situ Joule heating with Buckypaper (up to 177°C). Characterisation results demonstrated that composites fabricated by this out-of-autoclave curing method exhibited relatively good mechanical properties and low void content. In addition, finite element analysis indicated that this method had the advantages including uniform heat distribution and energy efficiency. Therefore, Buckypaper heating is a promising technique for composite curing with potential for energy & cost savings.

Copyright © SeaBioComp

Many researchers have investigated the application of integrally heated mould tool in composite production. Liu et al [28] reported an out-of-oven curing method by integrating a pyroresistive surface layer (based on graphene nanoplatelets and high density polyethylene) into a composite laminate. The developed surface layer possessed a positive temperature coefficient, which enables a separation of the conductive network when reaching at the predetermined temperature to prevent the overheating. It also had good flexibility for the manufacturing of structures with complex shapes. Compared to state-of-the-art out-of-autoclave oven curing, the proposed out-of-oven Joule heating approach consumed only 1% of the energy required for curing, with no effect on mechanical performance and glass transition temperature (T_g) of the final composite.

Abdalrahman et al [29] studied the design optimisation of an integrally-heated tool numerically using Taguchi methodology. It was found that the heating channel layout is the most crucial parameters for the thermal performance of the integrally-heated tool, and parallel layout proved to be the most efficient to achieve the desired characteristics of surface temperature uniformity and minimum heating time. In addition, channel cross-section showed very little influence on heating efficiency, while the heating channel separation need to be determined according to the production requirement. Bromley [30] investigated integrally-heated tooling and thermal modelling methodologies for the rapid cure of aerospace composites. He "recommended to use a reliable oven for the large majority of the part that does not need the active temperature control provided by integrally heated tooling and only use the localised heating control for the thickest portions of the laminate". He also said" heating a part from the underside of the tool face while utilising an insulation blanket over the top does appear to provide benefits over oven curing for certain part families, and can lead to faster cure times. The energy usage difference, although theoretically less, is not as great as anticipated, due to ovens being more efficient than previously thought". However, the analysis was for relatively small parts compared to those intended after the culmination of the SeaBioComp project.

3 Demonstrator components

Discussions on demonstrator components were held with two companies interested to use biocomposites in the marine environment.

Siemens Gamesa Renewable Energy (SGRE in UK and Germany) required a Non-Disclosure Agreement to discuss potential SeaBioComp demonstrator components. They proposed that the demonstrator might be either (a) a wind turbine spinner or (b) a wind turbine nacelle cover. The component is normally mounted at the top of the support tower and as such is sensibly remote from exposure to the full rigours of the marine environment.

JET Engineering is developing a network of 5G telecommunication buoys to provide mobile phone and data connectivity at sea [31-36]. The proposed system floats on a ~1.2 m diameter buoy with a domed housing for the telecommunications equipment surrounded by solar panels. In the first instance, the complex curvature of the dome provides an interesting challenge for "wiring" an electrically heated mould tool (and electrically heated silicone membrane counterface) and is chosen as the demonstrator component. The shape has similar characteristics to the spinner (nose cone) of a wind turbine without the added complexity of the blade apertures.

4 Conclusion

This report reviews the suitable materials for the mould tool and heating elements available on the market. Most importantly, the design of a 3D heated mould tool for an offshore wind turbine nacelle cover demonstrator is presented, and the achievement of sensible thermal uniformity has been investigated using an infrared camera as the temperature has a significant influence on the property of the final composite structure.

Copyright © SeaBioComp

Acknowledgement
The authors acknowledge useful discussions on demonstrator components with Christopher Briggs (Siemens Gamesa Renewable Energy (SGRE) ~ UK), Jeanette Haenseroth, Carlos Llopis (SGRE ~ Germany), Jonas Pagh Jensen, Harald Stetcher (SGRE ~ Denmark), Xabier Hierro-Olabarria Salgado, Naiara Doblas Telletxea (SGRE ~ Spain), R Ramkumar (SGRE), and with James Thomas (JET Engineering).

References

1. N Pearce, FJ Guild and J Summerscales, An investigation into the effects of fabric architecture on the processing and properties of fibre reinforced composites produced by resin transfer moulding. Composites Part A: Applied Science and Manufacturing, **1998**, *29(1-2)*, 19-27.
2. J Summerscales, Resin infusion under flexible tooling (RIFT). Wiley Encyclopedia of composites **2011**, 1-11.
3. C Williams, J Summerscales and S Grove, Resin infusion under flexible tooling (RIFT): a review. Composites Part A: Applied Science and Manufacturing**, 1996**, *27*, 517-524.
4. J Summerscales and T Searle, Low-pressure (vacuum infusion) techniques for moulding large composite structures. Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications, **2005**, *219*, 45-58.
5. Y Qin, J Summerscales, J Graham-Jones, M Meng and R Pemberton, Monomer selection for in situ polymerization infusion manufacture of natural-fiber reinforced thermoplastic-matrix marine composites. Polymers**, 2020**, *12*, 2928.
6. O Avinc and A Khoddami, Overview of poly (lactic acid)(PLA) fibre. Fibre Chemistry, **2010**, *42*, 68-78.
7. E Louisy, F Samyn, S Bourbigot, G Fontaine and F Bonnet, Preparation of glass fabric/poly (L-lactide) composites by thermoplastic resin transfer molding. Polymers, **2019**, *11*, 339.
8. MW Arney, SM Grove, I Progoulakis, TJ Searle, D Short, J Spooner and J Summerscales, Integrally-heated tooling for the manufacture of fibre-reinforced composites. In Proceedings of Composites Processing 2004, Composites Processing Association, Bromsgrove, 23 April 2004.
9. R Stewart, New mould technologies and tooling materials promise advances for composites. Reinforced Plastics**, 2010**, *54(3)*, 30-36.
10. Cytec Engineered Materials. Availabe online: https://www.cytec.com (now Solvay, https://www.solvay.com/en/, accessed on 10 March 2023.
11. The Advanced Composites Group. https://theadvancedcompositesgroup.com/, accessed on 10 March 2023.
12. Amber Composites. http://www.ambercomposites.com/, accessed on 10 March 2023.
13. GrafTech. https://www.graftech.com/home/default.aspx, accessed on 10 March 2023.
14. Touchstone Research Laboratory. https://www.trl.com/, accessed on 10 March 2023.
15. Fibretemp®. https://www.fibretemp.de/en/, accessed on 10 March 2023.
16. Tibgrid®: flexible heating fabrics and heating textiles for multilayers or composite parts and tooling, accessed on 10 March 2023.
17. Heated moulds and lightweight components made of fiber composites, https://cotexx.de/en/electrical-surface-heating-how-to-heat-composites, accessed 10 March 2023).
18. Progoulakis, L. Heated tooling for aerospace composites manufacture. MPhil thesis, University of Plymouth, 2004.
19. Y Ding, WK Chiu, X Liu and B Whittingham, Modelling of thermal response of oil-heated tools due to different flow rates for the manufacture of composite structures. Composite Structures, **2001**, *54*, 477-488.
20. Anon., Mould tool heating – the oven-free alternative. Reinforced Plastics**, 2003**, *47*, 38-41.
21. B Griffiths and N Noble, Process and tooling for low cost, rapid curing of composites structures. SAMPE Journal, **2004**, *40(1)*, 41-46.
22. About Quickstep. https://www.quickstep.com.au/about/, accessed on 10 March 2023.

Copyright © SeaBioComp

23. The synthesis and applications of Buckypaper, https://nanografi.com/blog/buckypaper-and-its-potential-applications/, accessed on 10 March 2023.
24. Q Xia, Z Zhang, Y Liu and J Leng, Buckypaper and its composites for aeronautic applications. Composites Part B: Engineering, **2020**, 108231.
25. M Tarfaoui, A El Moumen, M Boehle, O Shah, and K Lafdi, Self-heating and deicing epoxy/glass fiber based carbon nanotubes buckypaper composite. Journal of Materials Science, **2019**, *54*, 1351-1362.
26. A Zhang and Z Li, Thermal properties of buckypaper enabled polymer composites. In Proceedings of MATEC Web of Conferences; p. 03005.
27. N Nguyen, A Hao, JG Park and R Liang, In situ curing and out-of-autoclave of interply carbon fiber/carbon nanotube buckypaper hybrid composites using electrical current. Advanced Engineering Materials, **2016**, *18*, 1906-1912.
28. Y Liu, T van Vliet, Y Tao, JJ Busfield, T Peijs, E Bilotti and H Zhang, Sustainable and self-regulating out-of-oven manufacturing of FRPs with integrated multifunctional capabilities. Composites Science and Technology, **2020**, *190*, 108032.
29. R Abdalrahman, S Grove, A Kyte and MJ Rizvi, Numerical simulation and design optimisation of an integrally-heated tool for composite manufacturing. Materials & Design, **2014**, *64*, 477-489.
30. HS Bromley, Investigation of integrally-heated tooling and thermal modeling methodologies for the rapid cure of aerospace composites. Massachusetts Institute of Technology, 2015.
31. JET launches world's first 5G connected sea buoy. https://jet-eng.co.uk/jet-launches-worlds-first-5g-connected-sea-buoy/, accessed on 10 March 2023.
32. 5G connected sea buoy off Lulworth Cove. https://www.dorsetecho.co.uk/news/19433715.5g-connected-sea-buoy-off-lulworth-cove/, accessed on 10 March 2023.
33. Press Release: JET opens Plymouth office to support 5G maritime operations. https://www.jet-eng.com/news/2021/4/11/press-release-jet-opens-plymouth-office-to-support-5g-maritime-operations (no longer available)
34. Maritime tech company moves into Plymouth freeport's Oceansgate. https://www.business-live.co.uk/technology/maritime-tech-company-moves-plymouth-20382103, accessed on 10 March 2023.
35. 5G Surf Condition Monitoring System to be deployed in Dorset. https://www.excelerate-group.com/5g-surf-condition-monitoring-system-to-be-deployed-in-dorset/, accessed on 10 March 2023).
36. 5G Aquaculture Video Monitoring System Deployed In UK. https://industryeurope.com/sectors/technology-innovation/5g-aquaculture-video-monitoring-system-deployed-in-UK/, accessed on 10 March 2023.

Copyright © SeaBioComp