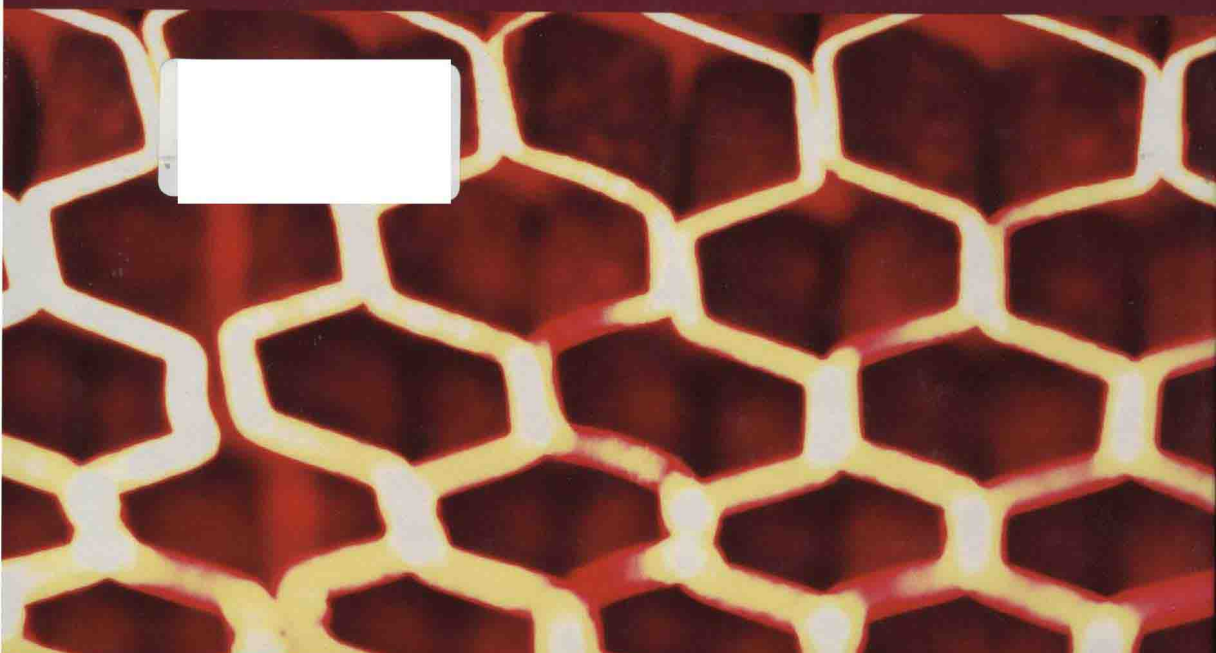


MECHANICAL ENGINEERING AND SOLID MECHANICS SERIES



Heat Transfer in Polymer Composite Materials

Forming Processes

**Edited by
Nicolas Boyard**

ISTE

WILEY

Series Editor
Noël Challamel

Heat Transfer in Polymer Composite Materials

Forming Processes



Edited by

Nicolas Boyard

ISTE

WILEY

First published 2016 in Great Britain and the United States by ISTE Ltd and John Wiley & Sons, Inc.

Apart from any fair dealing for the purposes of research or private study, or criticism or review, as permitted under the Copyright, Designs and Patents Act 1988, this publication may only be reproduced, stored or transmitted, in any form or by any means, with the prior permission in writing of the publishers, or in the case of reprographic reproduction in accordance with the terms and licenses issued by the CLA. Enquiries concerning reproduction outside these terms should be sent to the publishers at the undermentioned address:

ISTE Ltd
27-37 St George's Road
London SW19 4EU
UK

www.iste.co.uk

John Wiley & Sons, Inc.
111 River Street
Hoboken, NJ 07030
USA

www.wiley.com

© ISTE Ltd 2016

The rights of Nicolas Boyard to be identified as the author of this work have been asserted by him in accordance with the Copyright, Designs and Patents Act 1988.

Library of Congress Control Number: 2016931428

British Library Cataloguing-in-Publication Data

A CIP record for this book is available from the British Library

ISBN 978-1-84821-761-4

Heat Transfer in Polymer Composite Materials

Preface

This book is a recapitulation of the CNRS Summer School organized in June 2013 dedicated to heat transfer in organic matrix composite materials and their forming processes. The industry of composites has grown steadily in recent years due to the numerous advantages of these materials, such as their lightness and interesting mechanical properties, compared to aluminum and other metal-based alloys. To remain competitive, especially in a very strong international economic context, the quality of the produced parts must be fully controlled. This control requires an accurate knowledge of physical phenomena occurring during the various steps of their manufacturing process and in a context where the strong activity and the needs led to the emergence of new processes and increasingly fast production rates. The forming of composite materials has thus become a major topic of research in terms of experimentation, modeling and simulation, where several scientific disciplines must come together in order to achieve the control of manufactured parts and properties. We can notice that heat transfer is one of the main levers to control the forming processes and induced properties of the composite part. They have to be carefully analyzed during the manufacturing of these materials that also require a multidisciplinary approach. Thus, thermal sciences have to be coupled to other scientific fields such as mechanics and physical chemistry.

The first goal of this summer school was to bring together academic and industrial researchers from different disciplines within thermal sciences with transverse themes common to their activities. A second aim was also to provide the basis on heat transfer during polymer and composite processing as well as the latest methods and techniques from experimental, numerical and modeling points of view, useful to help in the solving of many issues. Therefore, the book takes this and gives theoretical and practical information to understand, measure and describe, in a relevant way, heat transfer during forming processes (in the tool as well as in the composite part) and introducing the required couplings. For this purpose, we relied on the experience of recognized French researchers.

This book is written in a comprehensive way for an audience that is already aware of the world of composites and associated processes: graduate students, researchers and people involved in R&D activities in industrial sectors. Our aim is to provide a tool, useful for the readers to start a study on composite processing where heat transfers are involved. Each chapter describes the concepts, techniques and/or models related to the developed topic and several examples are given for illustration purposes. A list of selected references is also given at the end of each chapter for a deeper complement of its content, which is necessary for more complex analyses and developments. Unfortunately, all topics and issues related to heat transfer in composite parts and processes cannot be addressed in a single book and a selection was made to cover a broad range of subjects and associated issues.

The introductory chapter presents heat transfer analyses and issues in polymer and composite processing through illustrative examples mostly from injection molding. Preconceived ideas, difficulties and simplified approaches are well highlighted. One key to success in heat transfer modeling is the accurate knowledge of thermophysical properties, phase change kinetics and their associated models for both thermosets and thermoplastics. Conventional as well as new methods to experimentally determine these properties and reaction rate parameters as a function of temperature are detailed in Chapters 2 and 3. Scientific and technical issues are also included. A comprehensive review of the effects of thermoplastics process conditions (shear and/or elongation induced by the flow) and the addition of other components (nucleating agents, fibers, etc.) on the transformation kinetics of the polymers, their rheological behaviors and final microstructures is detailed.

From all these data, the simulation of residual stresses developed during the matrix transformation and the cooling is discussed. For this purpose, thermokinetic and mechanical couplings are introduced and the prediction of cure-dependent mechanical properties is presented. In Chapter 6, modeling of heat transfer in multi-scale porous media, which can be encountered during the filling step of Resin Transfer Molding (RTM) mold, is discussed following a homogenization approach. The relationship between the physics at local-scale and the macroscale description is explained, also including the determination of effective properties.

The improvement of the quality of parts can be achieved by optimizing process parameters. Among them, the thermal control of the part is of strong importance and depends on the thermal control of the mold. Thus, optimization approach has to consider heat transfer in the tool and couplings to include contact conditions and transformation kinetics. Context, definition and methods of optimization are covered in this book and are illustrated with two detailed examples. Process modeling is introduced in Chapters 8 and 9. First, we discuss the peculiar case of thermoplastic welding, where no adding materials is required for assembling. The importance of intimate contact and macromolecular diffusion is emphasized from theoretical and

practical points of view, including the strong temperature dependence. A simulation of forming processes is also addressed in a more general way. Several examples are proposed to present multiscale, multiphysics and multidomain modeling, which are representative of the complexity of forming processes.

Another important part in heat transfer analysis concerns the instrumentation for the thermal characterization and the control of manufacturing processes. From these data, it is possible to obtain information about the process, thermophysical properties and/or the matrix transformation (for example, using inverse method algorithms). An overview of the existing instrumentation (contact and contactless methods) is given in this book. A specific chapter has been specifically dedicated to heat flux sensors, since they provide relevant information to quantify heat transfer between the part and the tool. It is thus an important complement to temperature measurement. Available heat flux sensor technologies and their main characteristics are also mentioned and are completed with practical examples.

Toward the final part of this book, radiative heat transfer in polymer and composite forming are detailed. The processes using infrared heating are in development and the complexity of heat transfer analysis leads to several scientific issues. After a presentation of the basics to define thermal radiative properties, measurements are presented for classical semi-crystalline polymers and associated composites. Finally, after a description of infrared emitters and the temperature measurement using infrared camera, modeling of radiative heat transfer is introduced and polymer processing applications are included.

I would like to thank all my French colleagues who have done me the honor of participating to the CNRS Summer School in 2013 and then of accepting to contribute to this book project with their high-quality work. Special recognition goes to Didier Delaunay, CNRS senior researcher, for his scientific involvement and significant contribution to research in heat transfer in composites and forming processes. I hope that all readers, working in the broad field of polymer and composite processing, may find this book an interesting and valuable resource.

Nicolas BOYARD
January 2016

Contents

Preface	xv
Chapter 1. Introduction to Heat Transfer During the Forming of Organic Matrix Composites	1
Didier DELAUNAY	
1.1. Introduction.	1
1.2. Examples of injection of short fiber reinforced composites	2
1.2.1. Heat transfer during the filling phase	2
1.2.2. Heat transfer during part consolidation	18
1.3. Injection on continuous fiber reinforcements	22
1.4. Conclusion: toward a controlled processing.	24
1.5. Bibliography	25
Chapter 2. Experimental Determination and Modeling of Thermophysical Properties	29
Nicolas BOYARD and Didier DELAUNAY	
2.1. Measurement of specific volume and shrinkage	29
2.1.1. Thermoplastic PvT diagram.	30
2.1.2. Specific volume of thermosetting polymers	36
2.2. Determination of specific heat capacity of resin and composites	40
2.3. Thermal conductivity: a tricky task.....	43
2.3.1. A first assessment of experimental data.	44
2.3.2. Overview of the main characterization techniques to measure the thermal conductivity of polymers and associated composites	46
2.3.3. Modeling of the thermal conductivity of composites	57

2.4. Conclusions.	69
2.5. Bibliography	70

Chapter 3. Experimental Determination and Modeling of Transformation Kinetics	77
Nicolas BOYARD, Jean-Luc BAILLEUL and M'hamed BOUTAOUS	

3.1. Introduction.	78
3.2. What are the most suitable devices to analyze a reaction rate?	79
3.2.1. Conventional methods	79
3.2.2. Original methods	86
3.2.3. A first assessment of the current characterization methods	86
3.3. Modeling of the cure kinetics of thermosetting resins.	88
3.3.1. Mechanistic models: complexity versus accuracy.	89
3.3.2. Description of the kinetics with empirical models: the engineer approach.	91
3.3.3. Modeling of the diffusion induced by vitrification	97
3.4. Overall crystallization kinetics of semi-crystalline thermoplastics	98
3.4.1. Most popular crystallization kinetics models for process simulations.	100
3.4.2. Systems of differential equations.	102
3.4.3. Measurements of crystallization kinetics and associated parameters.	104
3.4.4. Models for specific crystallization phenomena and geometries	107
3.5. Concluding remarks.	109
3.6. Bibliography	111

Chapter 4. Phase Change Kinetics within Process Conditions and Coupling with Heat Transfer.	121
M'hamed BOUTAOUS, Matthieu ZINET, Nicolas BOYARD and Jean-Luc BAILLEUL	

4.1. Introduction.	121
4.2. Flow-induced crystallization: experimental observations.	124
4.2.1. Relevant experimental techniques	124
4.2.2. Effect of flow on crystallization kinetics	125
4.2.3. Flow effect on crystalline morphology	131
4.2.4. Flow effect on crystalline growth rate.	134

4.2.5. Effect of flow on rheological properties	134
4.2.6. Summary of experimental observations and guidelines for modeling	136
4.3. Flow-induced crystallization: modeling	138
4.3.1. Overall kinetics modeling	138
4.3.2. Explicit nucleation and growth modeling.	139
4.3.3. Role of viscoelasticity	140
4.4. Effect of the composite components	143
4.4.1. Effect of nucleating agents	143
4.4.2. Effect of fibers	144
4.5. Concluding remarks	147
4.6. Bibliography	149

Chapter 5. From the Characterization and Modeling of Cure-Dependent Properties of Composite Materials to the Simulation of Residual Stresses

Yasir NAWAB and Frédéric JACQUEMIN

5.1. Introduction.	157
5.2. Origin of residual stress	157
5.2.1. Mechanical levels of residual stress	158
5.2.2. Parameters contributing to the formation of residual stress	159
5.2.3. Problems generated by residual stress.	161
5.3. Determination of composite properties	161
5.3.1. Modeling the mechanical properties of composites.	162
5.3.2. Experimental determination of thermomechanical properties of composite	164
5.4. Modeling of residual stress.	167
5.4.1. Linear approach or classical theory of laminates	168
5.4.2. Nonlinear approach.	168
5.4.3. Minimization of energy approach	169
5.4.4. Application	170
5.5. Conclusion	171
5.6. Bibliography	172

Chapter 6. Heat Transfer in Composite Materials and Porous Media: Multiple-Scale Aspects and Effective Properties.

Michel QUINTARD

6.1. Introduction.	175
6.2. Effective thermal conductivity	177
6.2.1. Background on upscaling methods.	178

6.2.2. A simple example: continuous thermal conductivity	179
6.2.3. Effective thermal conductivity: properties and bounds.	181
6.3. Local-equilibrium model and thermal dispersion.	184
6.4. Local equilibrium versus local non-equilibrium models	187
6.4.1. The two-equation model.	187
6.4.2. Further discussion	189
6.5. Various extensions	192
6.5.1. Effect of homogeneous and heterogeneous thermal sources	193
6.5.2. Interfacial thermal resistance	194
6.6. Conclusions.	195
6.7. Bibliography	196

Chapter 7. Thermal Optimization

of Forming Processes	203
---------------------------------------	------------

Vincent SOBOTKA

7.1. Context of optimization.	203
7.2. Heat transfer: optimization lever	204
7.3. Definition of the optimization criterion	206
7.4. Problem modeling	207
7.4.1. Spatial scale	207
7.4.2. Time scale: process steps	207
7.4.3. Multi-physical aspects	207
7.5. Numerical optimization methods	208
7.5.1. The adjoint problem	210
7.5.2. Practical setting of the method	211
7.6. Example of process optimization: determination of optimal heat flux setpoint.	211
7.6.1. Experimental setup and constraint	212
7.6.2. Instrumentation of the mold and the preform	214
7.6.3. Thermal modeling	215
7.6.4. Experimental data from a composite part molding	218
7.6.5. Estimation of the thermal contact resistances	219
7.6.6. Determination of the optimal setpoint.	220
7.7. Optimal design of molds	222
7.7.1. OSOTO project	222
7.7.2. The considered thermoplastic part	222
7.7.3. General methodology	223
7.7.4. Conformal cooling approach	223

7.7.5. Heat transfer model in the process	224
7.7.6. Objective function	226
7.7.7. Minimization of the functional J	227
7.7.8. Cooling channel design	228
7.8. Conclusions and outlook	231
7.9. Bibliography	232

Chapter 8. Modeling of Thermoplastic Welding 235

Gilles REGNIER and Steven LE CORRE

8.1. Introduction.	235
8.1.1. Polymer welding processes	235
8.1.2. Healing mechanisms of polymer interfaces	237
8.2. Physics of thermoplastic welding	237
8.2.1. Intimate contact at interface.	237
8.2.2. Macromolecular diffusion.	241
8.3. Linear viscoelasticity to quantify the macromolecular diffusion	246
8.4. Application to continuous welding of composite tape.	248
8.4.1. Process description	248
8.4.2. Influence of processing conditions on interfacial strength	249
8.4.3. Modeling of macromolecular diffusion	249
8.4.4. Modeling of thermal aging	251
8.4.5. Thermal modeling of the process	253
8.4.6. Weldability prediction	254
8.5. Application to ultrasonic welding.	255
8.5.1. Process description and time scale separation	255
8.5.2. Process modeling: necessity of a time homogenization framework.	256
8.5.3. Numerical multi-physical model	257
8.5.4. Ultrasonic welding with energy directors: process analysis and optimization	259
8.6. Conclusion	262
8.7. Acknowledgments.	263
8.8. Bibliography	263

Chapter 9. Multiphysics for Simulation

of Forming Processes 269

Luisa SILVA, Patrice LAURE, Thierry COUPEZ and Hugues DIGONNET

9.1. Introduction.	269
9.2. Multiscale, multiphysics and multidomain modeling	270
9.2.1. Flow equations	271

9.2.2. Thermal-rheological-kinetical coupling	273
9.2.3. Orientation and structure development during processing	275
9.3. Advanced numerical techniques and macroscopic simulations	278
9.3.1. Implicit boundaries	280
9.3.2. Immersed subdomains and regularization	281
9.3.3. Multiphase flow and thermokinetical numerical resolution	282
9.3.4. Composite forming simulation illustrations	283
9.3.5. Parallel mesh adaptation and high- performance computing	286
9.4. Determination of equivalent properties and microscale simulations	289
9.4.1. Generation of representative numerical samples	289
9.4.2. Permeability of a composite	291
9.4.3. Stiffness tensor determination	294
9.5. Conclusions	294
9.6. Bibliography	296

**Chapter 10. Thermal Instrumentation for the
Control of Manufacturing Processes of Organic
Matrix Composite Materials**

Jean-Christophe BATSALE and Christophe PRADERE

10.1. Introduction	301
10.2. Methods based on contact measurement	302
10.2.1. Temperature sensors and fluxmeters	302
10.2.2. Heat flux estimation	304
10.2.3. Thermal probes for thermophysical property measurement in static conditions	308
10.3. Contactless heating	311
10.3.1. Photothermal methods with monosensors (under microscale characterization)	312
10.3.2. Thermal non-destructive evaluation: cracks or delamination detection in composite samples	313
10.3.3. Screening of chemical processes and microfluidic experiments	317
10.3.4. Principles for the factory global monitoring, example of conveyor belt parameter estimation	323
10.3.5. In a near future: the Big Data related to multiscale process survey	325

10.4. Conclusion	326
10.5. Bibliography	326

Chapter 11. Sensors for Heat Flux Measurement 333

Fabien CARA and Vincent SOBOTKA

11.1. Motivations: heat flux sensor	333
11.2. Principle of heat flux sensors.	335
11.2.1. Gradient heat flux sensor.	335
11.2.2. Inertial heat flux sensor	338
11.2.3. Inverse heat flux sensor	338
11.3. Main characteristics of HFS	340
11.3.1. Invasiveness	340
11.3.2. Time constant	340
11.3.3. Calibration	343
11.4. Type, positioning and use of heat flux sensors	344
11.4.1. Commercial sensors	345
11.4.2. Positioning of heat flux sensors.	347
11.4.3. Price	348
11.5. Advantages and limitations of HFS compared to other <i>in situ</i> monitoring techniques	349
11.5.1. Advantages.	349
11.5.2. Limitation and care in using HFS.	349
11.6. Examples	349
11.6.1. Compression molding	350
11.6.2. Resin transfer molding flow front detection	352
11.6.3. Resin transfer molding: influence of mold temperature	353
11.6.4. Internal temperature prediction during infusion	354
11.6.5. Glass mat transfer, internal temperature monitoring	355
11.7. Conclusions	356
11.8. HFS suppliers.	356
11.9. Bibliography	357

Chapter 12. Thermal Radiative Properties of Polymers and Associated Composites 359

Benoit ROUSSEAU

12.1. Introduction	359
12.2. Fundamental requisites concerning thermal radiation	361
12.2.1. Spectral range of thermal radiation.	361

12.2.2. Radiant energy, radiant flux, radiative flux density and radiative intensity	363
12.2.3. Blackbody spectral emissive power	366
12.2.4. Radiative properties at interfaces	368
12.2.5. Radiative properties of semi-transparent slabs	372
12.3. Prediction of the radiative properties of homogeneous semi-transparent slabs: case of the isotactic polypropylene.	373
12.3.1. Intrinsic optical properties for a homogeneous, isotropic and non-magnetic medium	374
12.3.2. Prediction of the radiative properties of polypropylene slabs at 20°C.	376
12.4. Radiative properties of polymer composites with fiber structures.	378
12.4.1. Normal spectral absorptance of Roving TWINTEx®	378
12.4.2. Normal spectral absorptance of sheet of PEEK with carbon fibers	380
12.5. Conclusion	381
12.6. Bibliography	381

**Chapter 13. Infrared Radiation Applied to
Polymer Processes 385**
Yannick LE MAOULT and Fabrice SCHMIDT

13.1. Introduction	386
13.1.1. Why use infrared heating for polymers?	386
13.1.2. Application of radiative transfers in polymer processing.	386
13.2. Infrared radiation characteristics.	388
13.2.1. Radiative properties (basis and main definitions)	389
13.2.2. Infrared emitters: characterization	393
13.2.3. Infrared camera measurements	397
13.3. Modeling of infrared radiation	403
13.3.1. Opaque medium: surface to surface methods.	403
13.3.2. Semi-transparent medium	405
13.3.3. Ray tracing method	407
13.4. Polymer processing applications.	409
13.4.1. Optimization of preform temperature for the ISBM process	409
13.4.2. Optimal infrared composite curing.	415
13.5. Future work	419