

Dr. Laure Moretti (P. D.)¹, Faheem Muhammed (Ph. D.M.S.E.)³, Tania Lavaggi (Ph. D.M.E.)², Prof. Suresh G. Advani^{1,2}, Prof. John W. Gillespie Jr.^{1,2,3,4}, Dr. Dirk Heider^{1,2}, Prof. Mark Mirotznik^{1,4}

University of Delaware | Center for Composite Materials¹ | Department of Mechanical Engineering² | Department of Materials Science and Engineering³ | Department of Electrical Engineering⁴

Introduction

Carbon/Carbon composites offer exceptional thermomechanical properties. However, they involve complex and expensive processes:

- High temperature / Long processing times
- Complex multi-physical phenomenon
- Risk of catastrophic failure...

To define and optimize these processes two approaches are possible:

✗ Long and expensive trial and error approach

✓ **Simulation**

The process studied is the Liquid Phase Infiltration using a thermosetting resin (see Fig. 1).

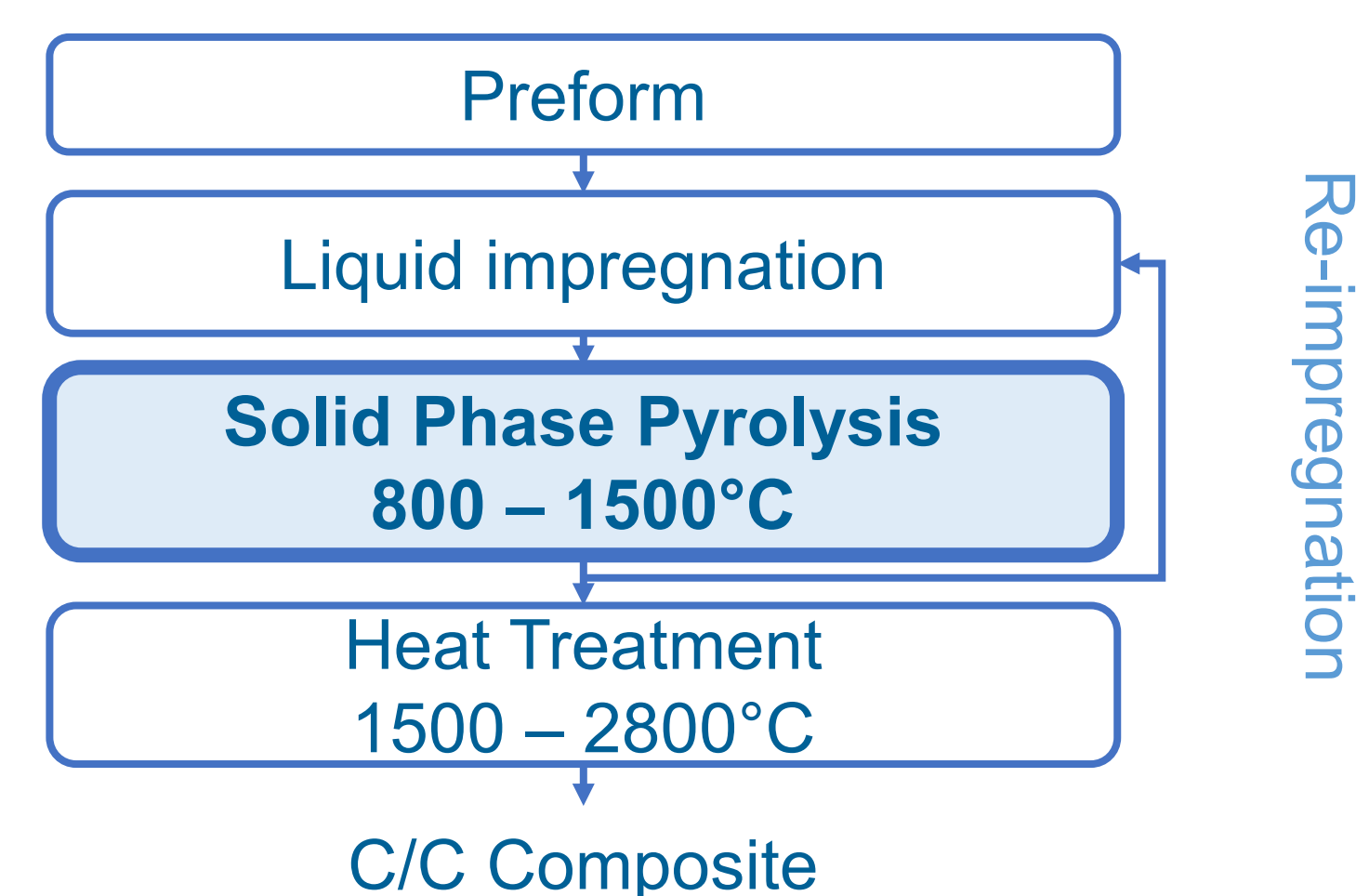


Fig. 1. Liquid Phase Infiltration - densification process

During pyrolysis, the material is submitted to high temperatures, the matrix experiences drastic changes of properties strongly impacting the porosity and permeability of the composite.

► Develop a tool to simulate the degradation of the matrix and to tailor the permeability and final properties of the composite

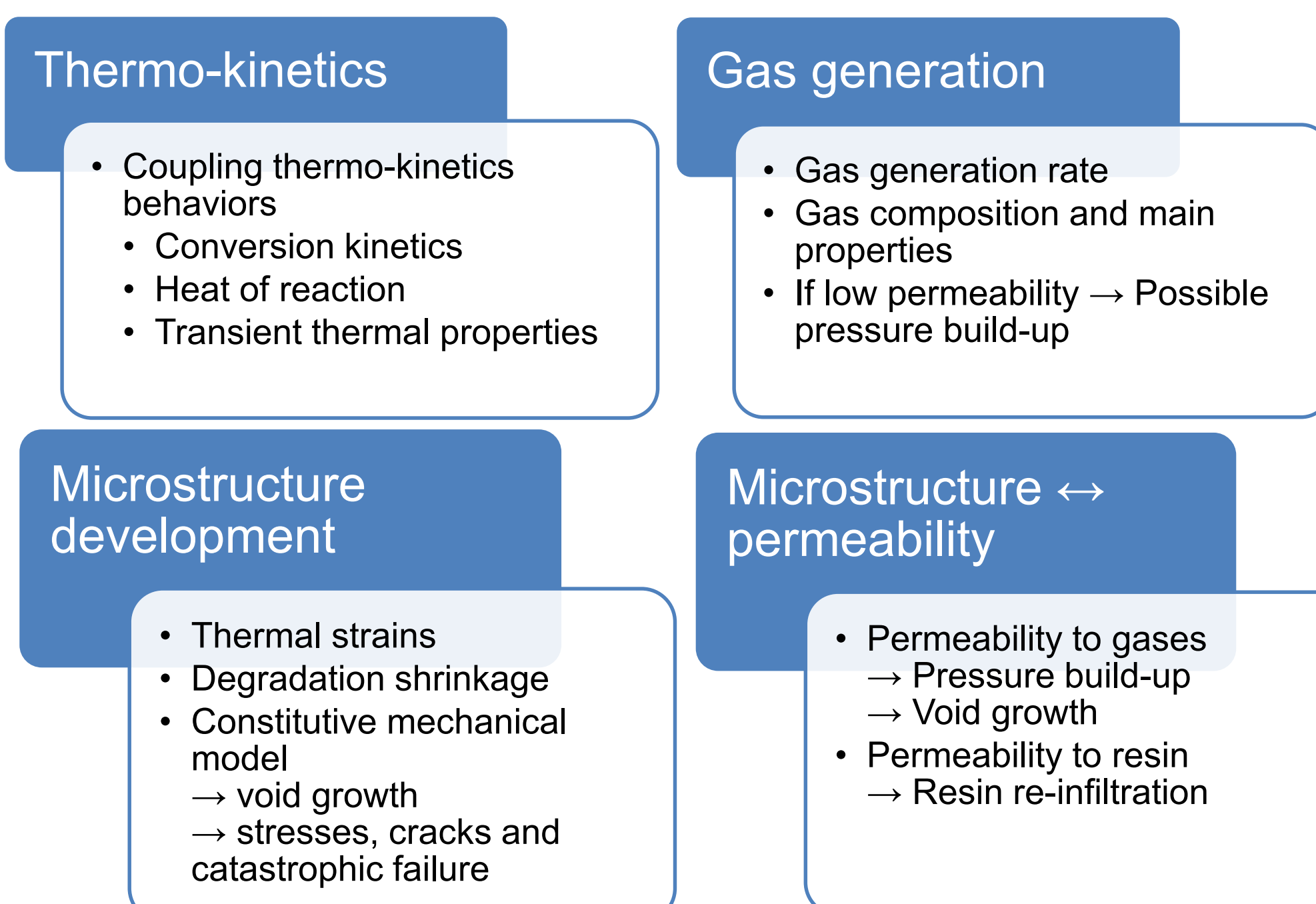


Fig. 2. Main multi-physical phenomenon

Pressure and Temperature

Numerical tool simulates:

- Pressure
- Temperature
- Transient material properties impacted by the temperature and associated kinetics

$$\alpha = \frac{M_0 - M}{M_0 - M_f}$$

To do so, we use a Finite Difference Numerical Method applied to a 2D geometry (see Fig. 3).

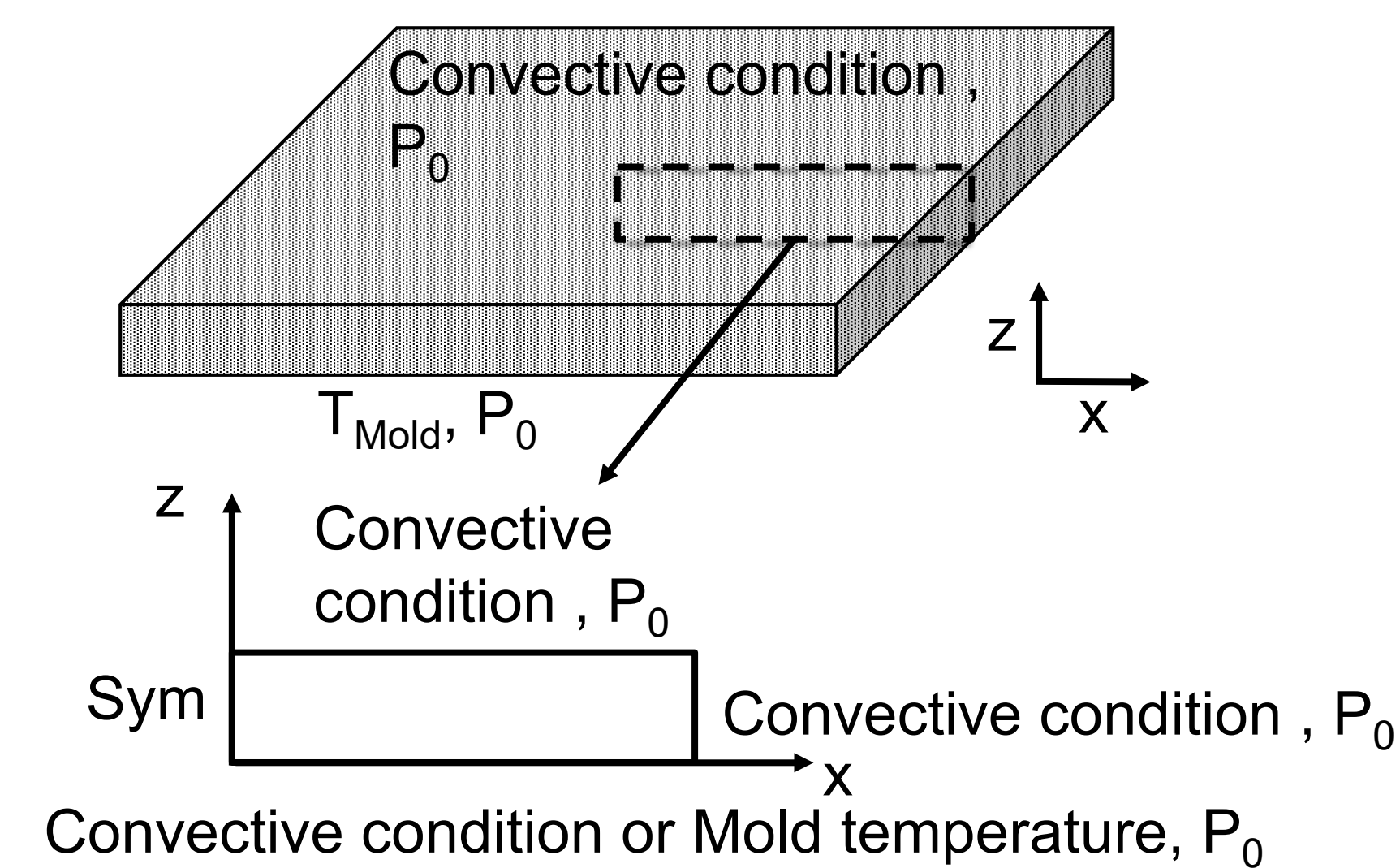


Fig. 3. 2D geometry

The results obtained for a benzoxazine based composite are described in Fig. 4. As the temperature increases and the matrix is converted, gases are released by the various reactions involved. Since the permeability of the material is initially very low, these trapped gases generate pressure.

While the degradation of the matrix progresses, the porosity and the number of cracks increase inside the composite creating new pathways for the gases to escape and increasing the permeability of the material. Eventually, most of the pressure inside the part is released as those gases are allowed to escape.

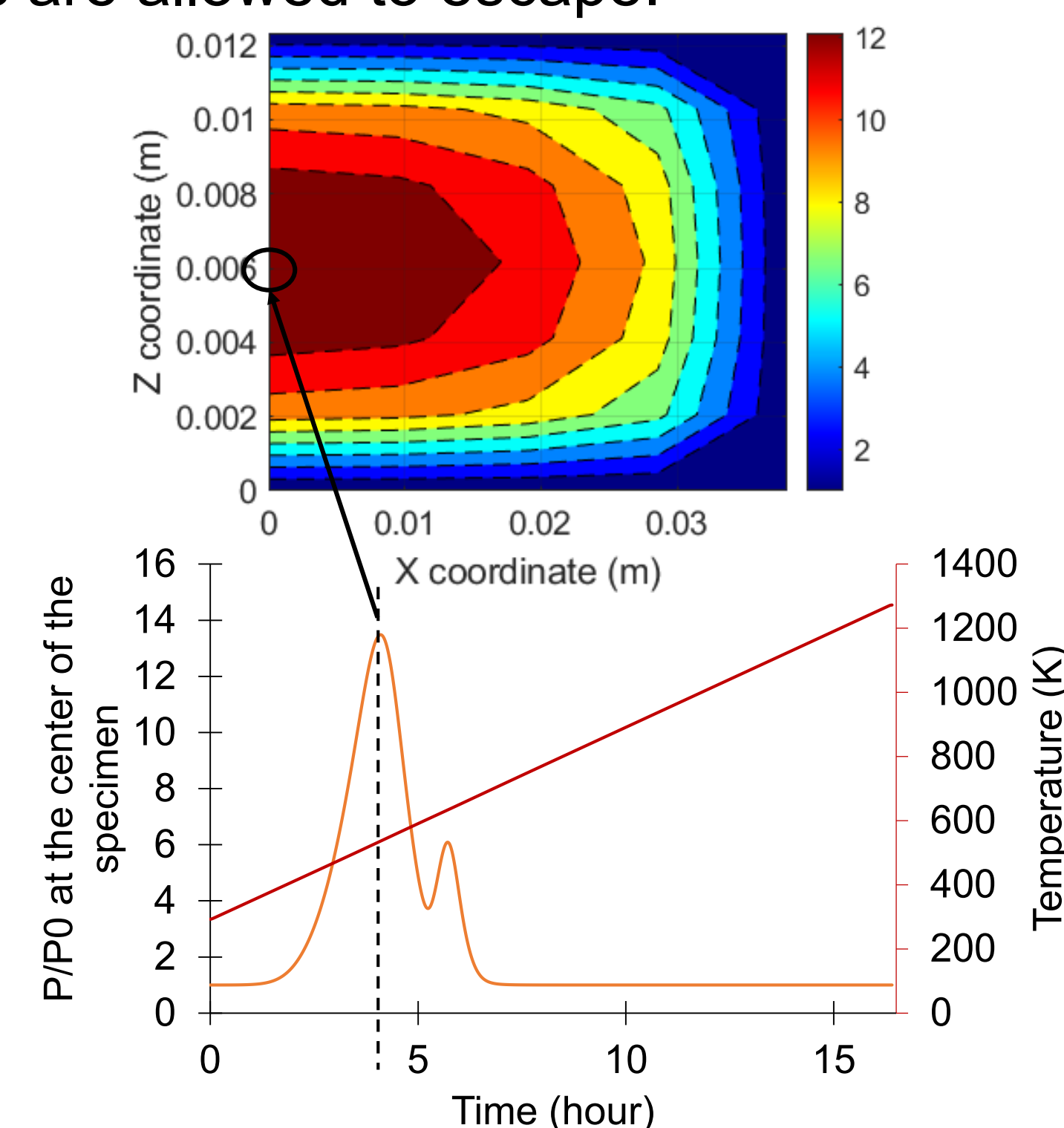


Fig. 4. Simulated pressure during the pyrolysis of a benzoxazine based composite (12 mm thick part)

Microstructure Development

Main mechanisms impacting the microstructure:

- Gases release and pressure increase
- Coefficient of thermal expansion (CTE) mismatch between fibers and matrix
- Degradation shrinkage of the matrix

As described by the numerical tool, when the temperature increases and the matrix is converted, gases are released

- Generation of gases and voids
- If low permeability → Pressure builds up
 - If matrix stiffness low enough → Pressure can expand voids
 - If matrix stiffness high enough → Stresses, cracks and, eventually, catastrophic failure

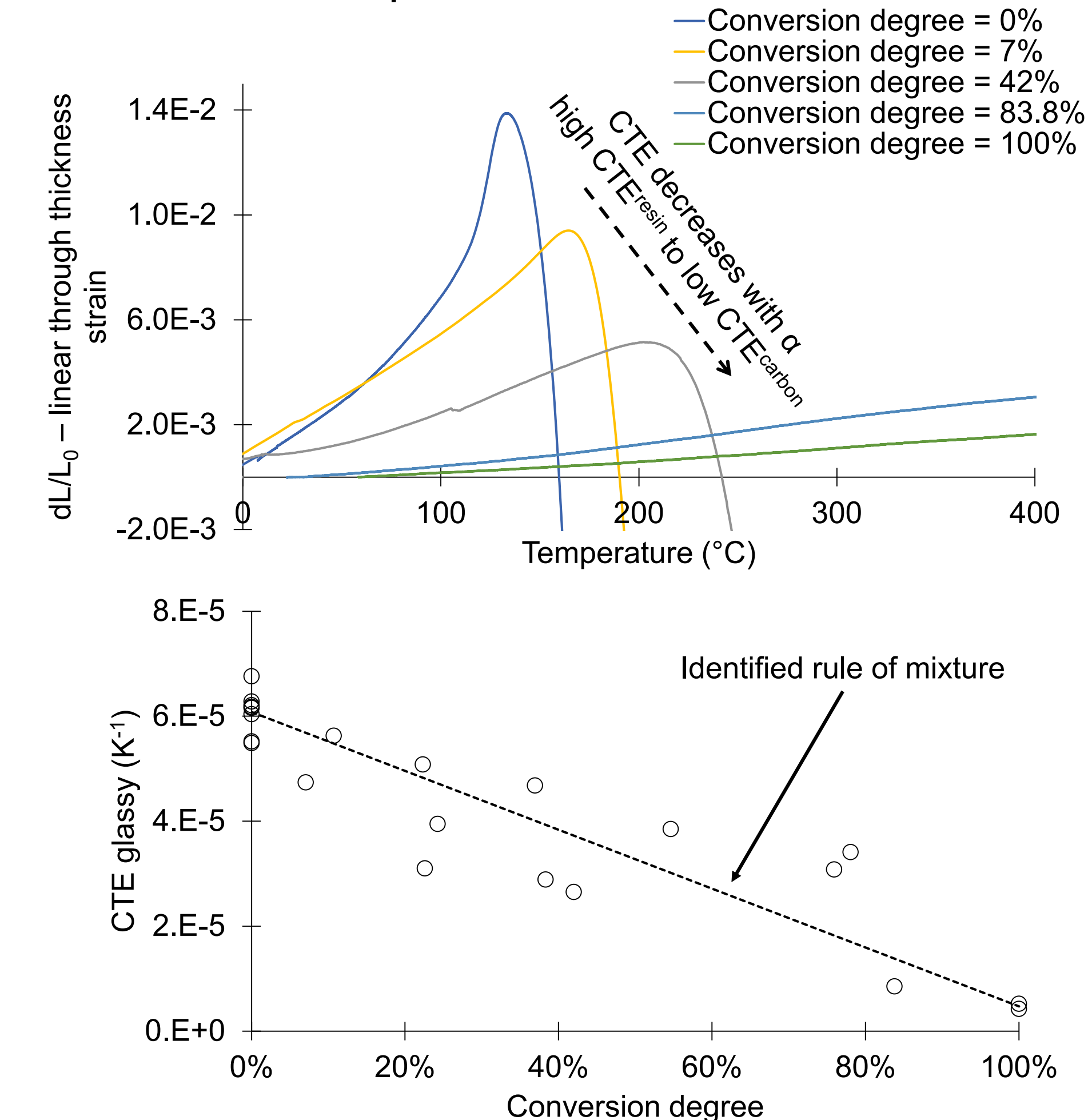


Fig. 5. Thermal strains and matrix CTE evolution during pyrolysis

- Thermal strains: CTE mismatch between the evolving matrix, the low transverse fibers CTE and the negative longitudinal fibers CTE
 - If matrix stiffness high enough → stresses and cracks can appear
- Matrix CTE evolution during pyrolysis: TMA measurements (see Fig. 5) on matrix (no fibers) specimens with different initial conversion degrees α
- Rule of mixture according to α :
 $CTE = (1 - \alpha) \cdot CTE^{cured} + \alpha \cdot CTE^{carbonized}$

- Degradation shrinkage strains: mismatch between the stable fibers and the shrinking matrix
- If matrix stiffness high enough → stresses and cracks

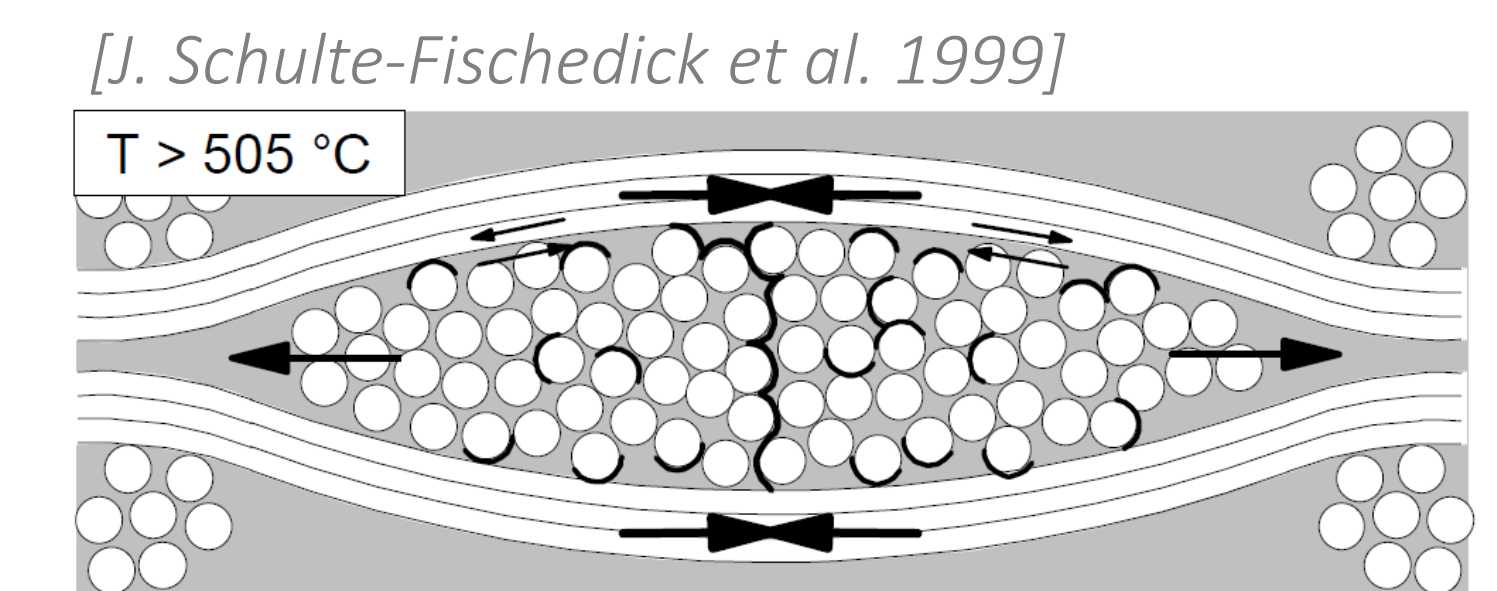


Fig. 3. Expected stresses during the main pyrolysis

Conclusions and Prospects

Steps of the research project:

- Characterization of the material behavior and the development of the microstructure
 - Characterization and identification of the behavior laws associated with the degradation shrinkage and density change experienced by the material at high temperatures
 - Characterization of the evolution of the glass transition temperature and the modulus of the matrix and identification of a constitutive mechanical model to be able to describe:
 - void growth
 - stresses, cracks, catastrophic failure
- Correlation between the evolution of the microstructure and the permeability of the material
- Simulation and optimization of the pyrolysis
 - Current numerical simulation
 - Pressure and temperature
 - Transient material behavior
 - In progress: void growth, stresses, cracks and resulting permeability
- Comparison with experimental microstructure, re-infiltration and validation

Acknowledgements

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