Self-Healing Thermosets and Composites: Status and Future Perspectives for the Industry

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Self-healing concept:

Two possible approaches for self-healing polymers

- **Extrinsic systems (Part I)**
  - **Healing agents** to repair damage
  - **Newly formed bonds** as thermally stable as the ones in the bulk
  - Applicable to multiple materials

- **Intrinsic systems (Part II)**
  - **Reversible bonds** in the bulk
  - Rupture along these chemically weaker bonds
  - **Facilitated multiple healing** at the same spot
  - Application specific systems

‘A self-healing material has the built-in ability to fully or partially repair the damage occurring during its life-time’
Extrinsic systems

- Potential healing agent reservoirs:
  a) Microcapsules
  b) Hollow fibers
  c) Carbon nanotubes
  d) Microvascular networks

- Healing agent reservoirs placed in matrix materials
  - Light and strong matrices
  - Epoxy, PE, vinylester, PU resins
  - Damage: Wear, degradation, microcracks…
1. Design of dedicated polymer-based healing agents for a range of thermoset materials and composites with good sealing & healing ability, also under harsh environmental conditions;

2. Design of suitable encapsulation technologies, including the design of a system suitable for multiple healing actions;

3. Design of chemical nature of thermoset matrix in such a way that an increased compatibility with the healing agents can be expected;

4. Design of a multi-step healing system, combining fast sealing and crack limitation, with a subsequent complete healing action;

5. Development of an integrated methodology framework around self-healing principles including modelling, fracture mechanics and kinetics.
SEPOCOM Team

Polymer science
Composite research
Non-destructive measurement techniques
Advanced thermal analysis
Durability & sustainability of structural materials

PCR (UGent, F. Du Prez)
UGCT (UGent, V. Cnudde)

MMS (UGent, J. Degrieck; W. Van Paepegem)

Numerical modeling

MeMC (VUB, D. Van Hemelrijck)

FYSC (VUB, H. Rahier)
Healing Chemistry

- **Reaction properties**
  - Fast inhibition of crack propagation
  - High yields
  - No release of (toxic) compounds
  - Suitable in a **wide range of conditions**

- **Selection of healing agents**
  - Liquid with high boiling point
  - Compatibility with matrix
  - Commercial availability or easy synthesis
  - High crosslinking density
  - **Stable at ambient conditions**
Healing Chemistry

- Online FT-IR spectroscopy
- Differential Scanning Calorimetry (DSC)

F.E. Du Prez et al., *Polymer*, 2012, 53, 2320

(VUB – FYSC, H. Rahier)
Microcapsules

Shell properties

- Mechanical properties:
  - **Brittle** to break while cracks propagate into the material
  - **Robust** to survive their manufacture

- Thermal stability

- Resistance to solvents

- **Good shelf life** to collect them as a powder and insert them into materials
Microcapsules

SEM pictures

Size & morphology can be controlled
Analysis and results

- **Healing through manual injection (TDCB-testing)**
  - Mixture of healing agents injected between the crack planes
  - Allowed to heal for 1/3/5 days at room temperature (25 °C)
  - Increase in *efficiency* over time: up to **130%** >>> previous systems

- **Healing with microcapsules**
  - 2 types of microcapsules inserted in epoxymatrix
  - *Efficiency* > **50%** after 5 days
  - Optimization needed

**Demonstrator:**
• Digital Image Correlation (DIC)
  Visualization of strain patterns

• Modeling
  Crack growth simulation with XFEM

(VUB – MeMC, D. Van Hemelrijck)

(UGent – MMS, W. Van Paepegem)
Crack propagation behavior

(UGent – MMS, W. Van Paepegem)
• X-Ray Computed Tomography (CT)
Non-destructive visualization technique

Distribution of capsules in matrix

(UGent – UGCT, V. Cnudde)
Intrinsic system: multi-step self-healing

Healing procedure:
- Shape memory recovery (< 2 min)
- Diels-Alder reaction (12h)
Multiple healing possible

scratched / healed at 50°C
Conclusions for intrinsic healing system

- **Tough and flexible** PU networks prepared in **one-pot** procedure
- Healing capability at **50°C**, based on shape memory + DA reversibility
- **Validated strategy** for micro/nano scratches (2D, 3D)
- **Recovery**: from a **structural** and **functional** point of view, in a **cyclic** way
- **Versatility**: applicable to multiple formulations