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3R-COMPO:
Reprocessable, Repairable and Recyclable Thermoset Composites for Aeronautics and Automotive

Nerea Markaide

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1. Why 3R-COMPO

Current limitations of thermoset composites:

1. Low-medium production rates
2. Recyclability
3. Repairability

These limitations are associated with the chemical process of resin crosslinking:

Solutions that are being investigated:

1. Reduction of crosslinking times
2. Reversible chemical bonds: Dynamic covalent chemistry

⬇

VITRIMER (Leibler, 2011)

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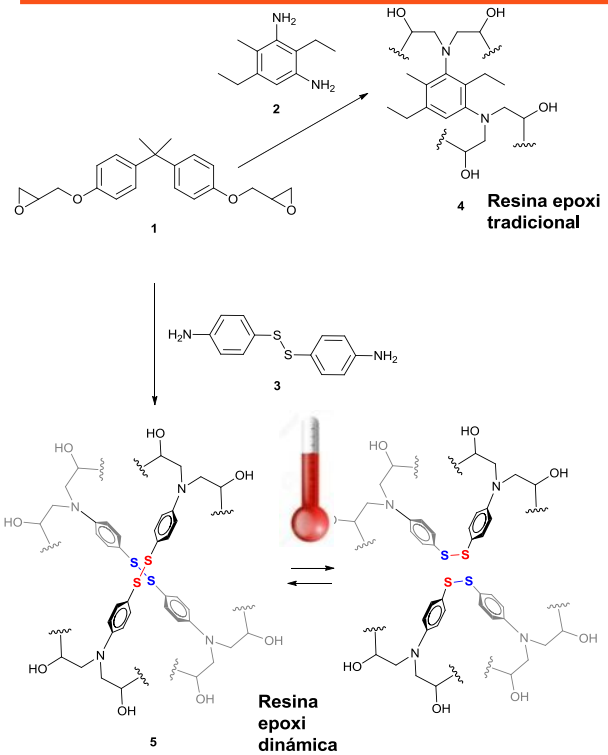
1. Why 3R-COMPO

Approach addressed in CIDETEC in 2013:

Dynamic epoxy system based on the exchange of aromatic disulfides:

- Simple synthesis
- Commercial raw materials



Hypothesis:

- When T < Tg: properties of thermoset resin
- When T > Tg+80ºC: Flexible, processability similar to thermoplastic resins.
- Therefore, it would provide the composite material with new functionalities:
  - **R**eprocesable
  - **R**ecyclable
  - **R**epairable

3R Concept

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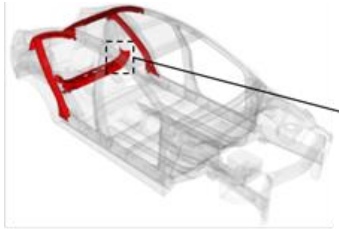
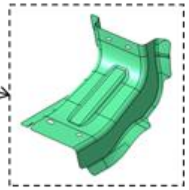
2. Project aproach

Thermoforming of cured flat laminate

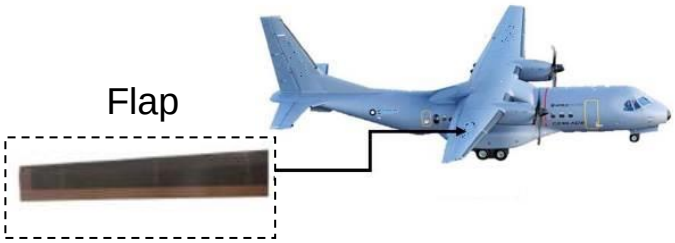
Reprocesability



Rear seat support



Flap



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3R-COMPO
(2015-2017)

Recyclability

Repairability

SEGULA TECHNOLOGIES

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AIMPLAS INSTITUTO TECNOLÓGICO DEL PLÁSTICO

Two possible ways:
- Chemical dissolution of resin
- Grinding and reprocessing

Repair of delaminations and resin microcracks applying P and T over damaged area

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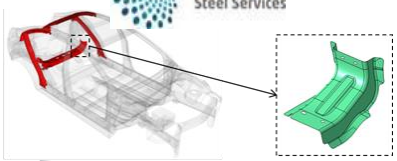
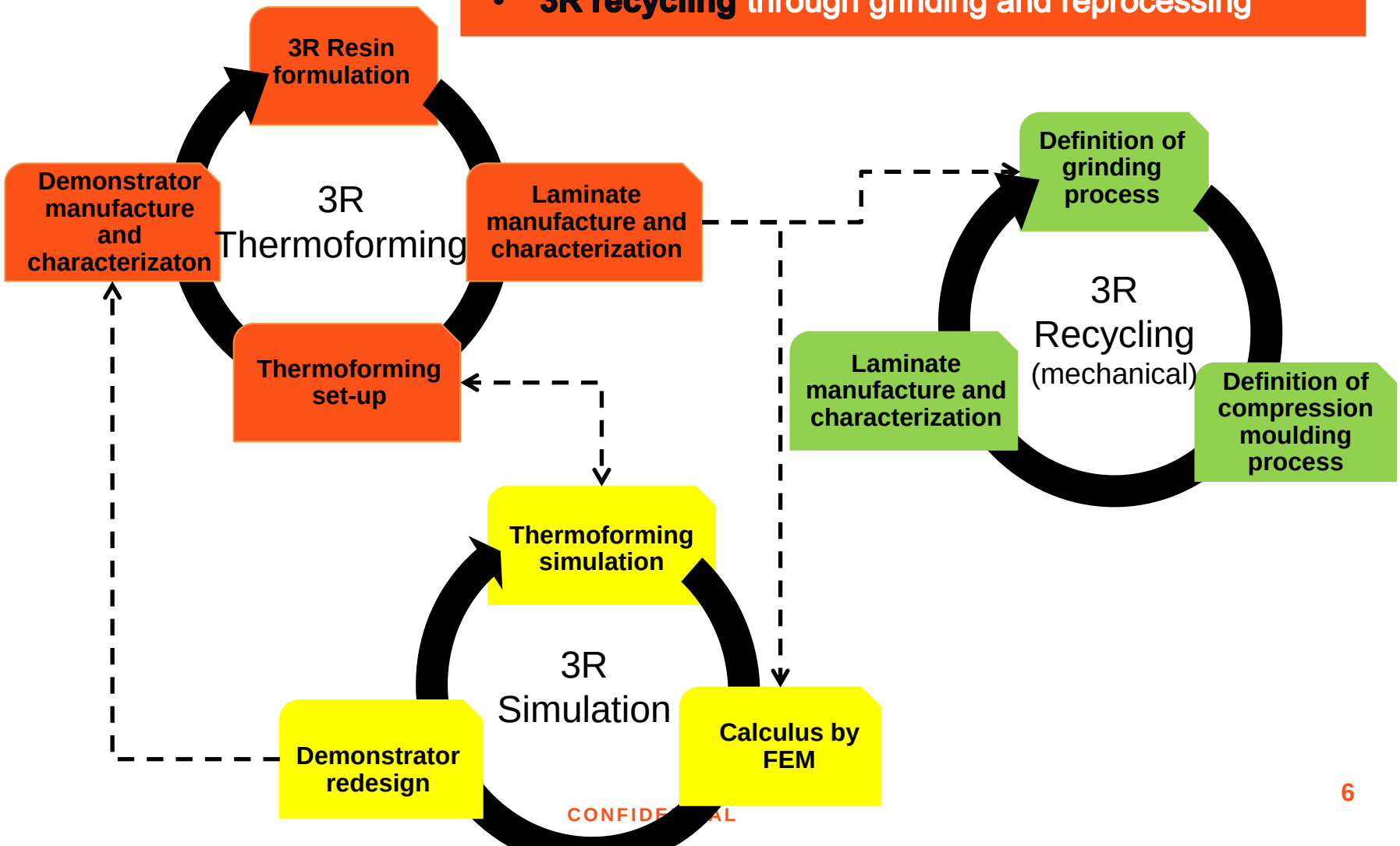


Objective:

- **3R laminate** and **3R simulation** to manufacture the demonstrator by **3R thermoforming**
- **3R recycling** through grinding and reprocessing

3. Experimental part and results



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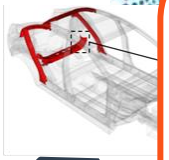
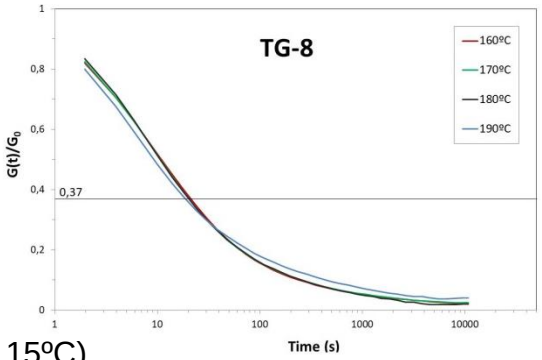
3. Experimental part and results



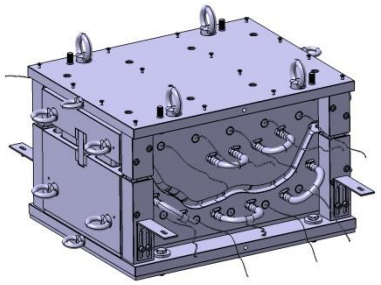
Study of different formulations.
Main aspects: viscosity, dynamic properties



DMA test: Evolution of the relaxation module when applying 1% of deformation on a cured specimen

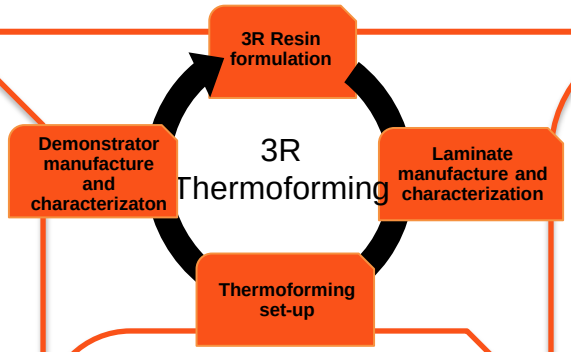
Epoxy resin: DGEBA for RTM
Dynamic hardener: 2-Aminophenyl disulfide
Curing cycle: 3 h @ 150ºC (105 < Tg < 115ºC)

Final mould and

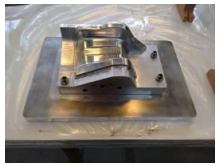


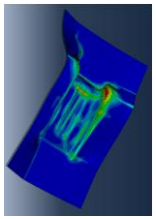
Thermoforming cycle:

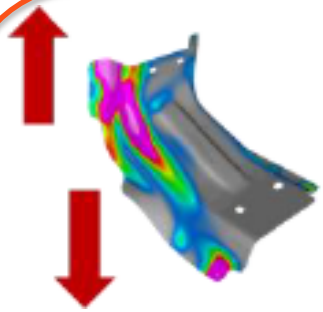
1. IR heating the laminate @ 210ºC
2. Compression @ 20-45 bar & 1 min
3. Cooling & demoulding below Tg



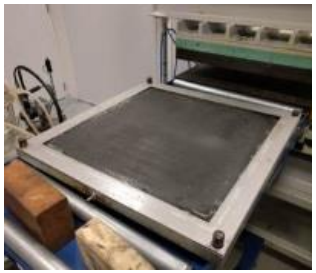
Scale mould and demonstrators supported by thermoforming simulation







FEM simulation



Laminate manufacture (RTM)

Mechanical properties	Standard	3R composite
Tensile Strength (MPa)	ISO 527-4	915±24
Young´s Modulus (GPa)		74,7±3,5
Flexural Strength (MPa)	ISO 14125	832±57
Flexural Modulus (GPa)		50,0±2,6

Carbon fabric: HS, 3K, twill 2x2, 200g/m^2
FVC: 50%

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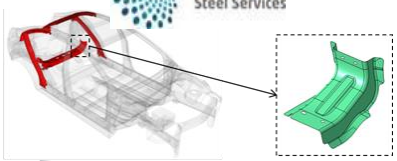
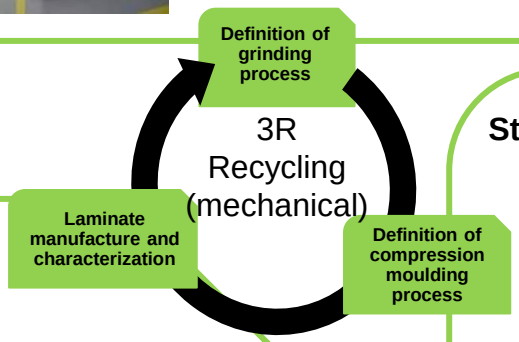


3. Experimental part and results

Study of grinding process and material size

Grinding machine

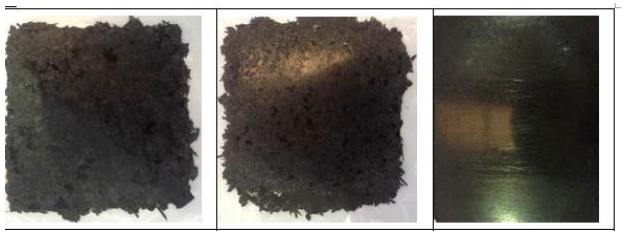
5-10 mm | 2-5 mm | <2 mm



Study of compression parametres

Optimum compression parametres:
200 bar @ 200ºC & 10 min

Characterization of laminates manactured with different grinding size



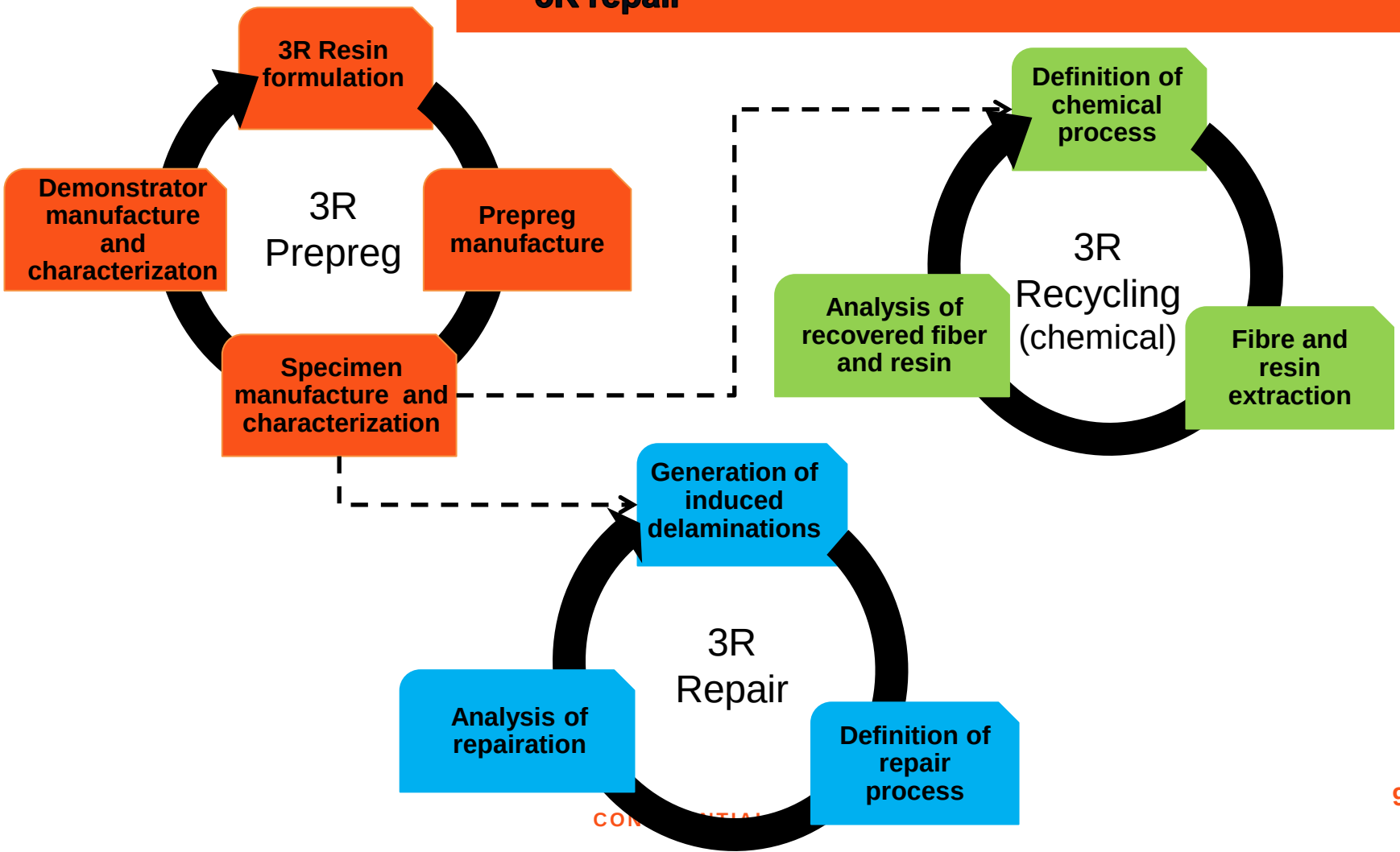
5-10 mm | 2-5 mm | <2 mm

Properties	Standard	3R composite		
		5-10 mm	2-5 mm	<2 mm
Flexural trength (Mpa)	ISO	108±13	95±8	126±8
Flexural Modulus (GPa)	14125	22,0±1,5	15,2±2,3	16,8±0,9

3. Experimental part and results

Objective:

- **3R prepreg** to manufacture the demonstrator by autoclave
- **3R recycling** through chemical dissolution of the resin
- **3R repair**



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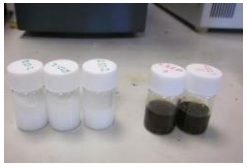
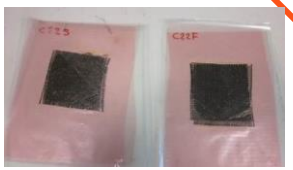
3. Experimental part and results

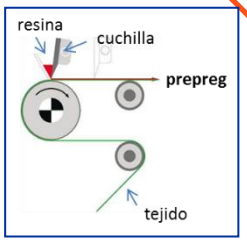
Study of different formulations.
Main aspects:
viscosity, flow, tackiness



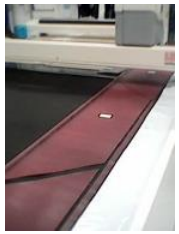


Epoxy resin: DGEBA for prepregging
Dynamic hardener: 4-Aminophenyl disulfide & 2-Aminophenyl disulfide
Curing cycle: 2,5 h @ 180ºC (130 < Tg < 140ºC)
2 h @125ºC (90 < Tg < 100ºC

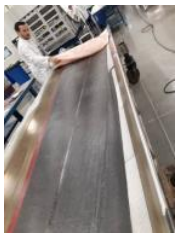
Prepregging machine



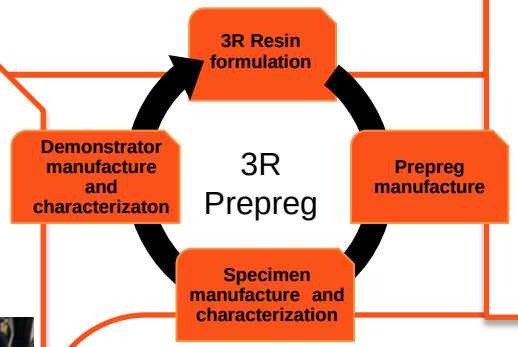
Knife system



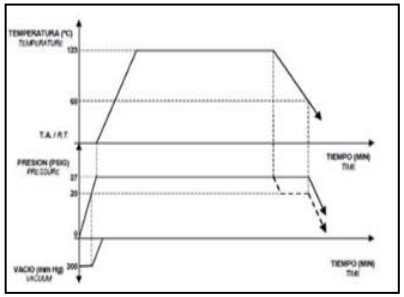
1. Ply cutting



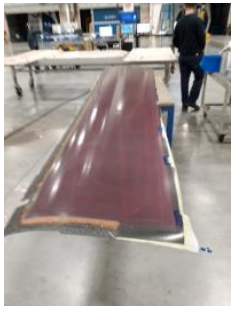
2. Lay-up



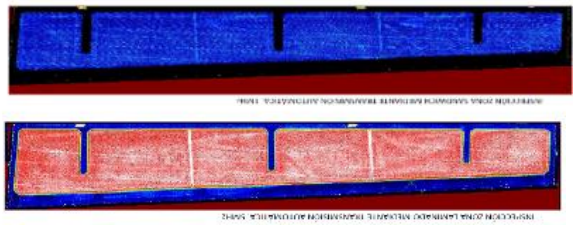
Details of impregnation and final prepreg roll



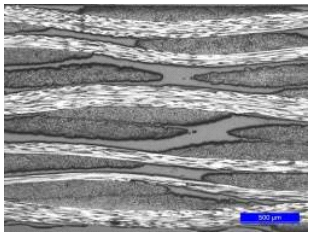
3. Curing in autoclave



4. Demoulding



5. NDT inspection



Carbon fabric: HT 3k 200 tex, plain 200g/m^2
FVC: 45%,

Properties	Standard	3R composite
Tensile Strength (MPa)	UNE-EN 2561	583,4±46,4
Compression Strength (MPa)	DIN-EN- 2850	140,0±6,7
Compression Modulus (GPa)		26,5±1,3
Interlaminar Shear Strength (MPa)	UNE-EN 2563	47,3±2,4
Fracture Toughness GIIC (KJ/m2)	ASTM D-7905	0,97±0,11

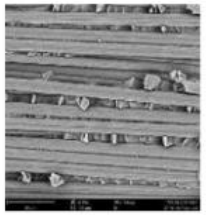
3. Experimental part and results

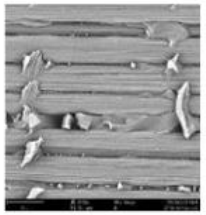
First fracture toughness test on 3R specimen

Properties	Standard	First test (After curing)
Fracture Toughness GIIC (KJ/m2)	ASTM D-7905	0,97±0,11

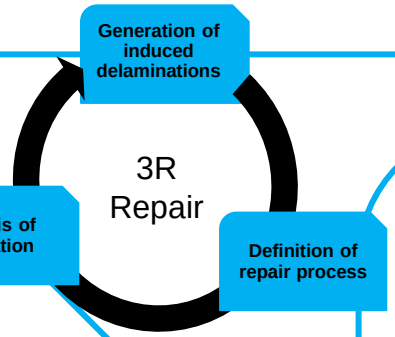
Carbon fabric: HT 3k 200 tex, plain 200g/m^2
FVC: 45%,


700x


2000x


5000x

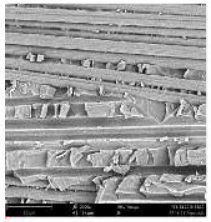
Fracture analysis by SEM

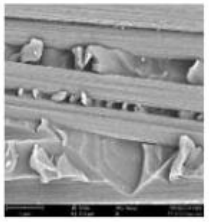


Second fracture toughness test on 3R specimen

Properties	Standard	Second test (After repairing)
Fracture Toughness GIIC (KJ/m2)	ASTM D-7905	0,97±0,15


700x

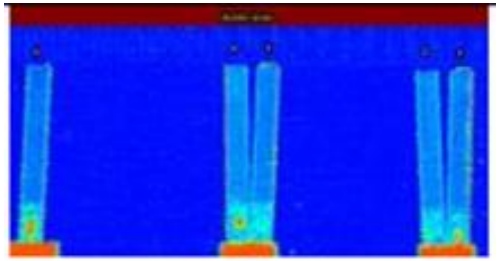

2000x


5000x

Fracture analysis by SEM

Press

Repair cycle: 10 min @ 200ºC & 200 bar



US inspection

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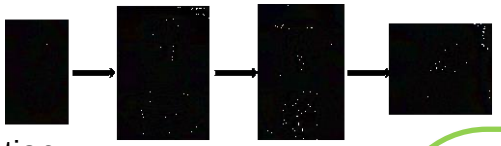
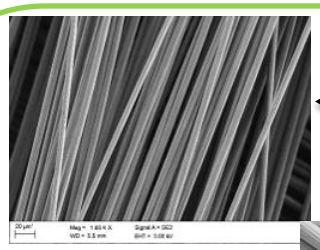
Experimental part and results

Study of the best chemical solution for resin disolution:

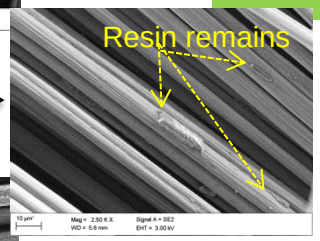


Chemical solution:

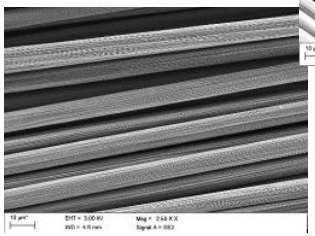
- Solvent
- Thiol (reductor agent)
- Catalyst



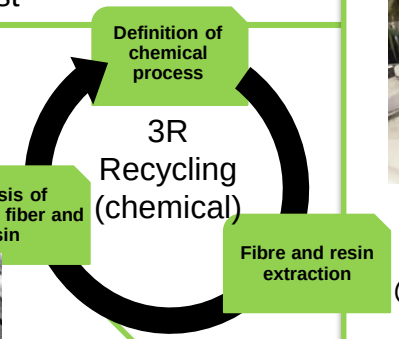
Virgin fibre



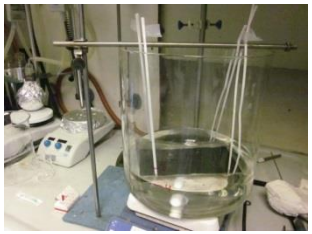
After 4 h in the chemical dissolution



After 4 h in the chemical dissolution (clean)

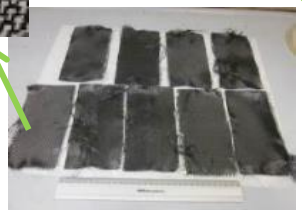


Scale-up



Laminate in the chemical solution @ 80ºC and stirring

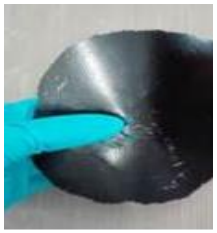
8 h



Recoverd fabrics

Dissolved resin

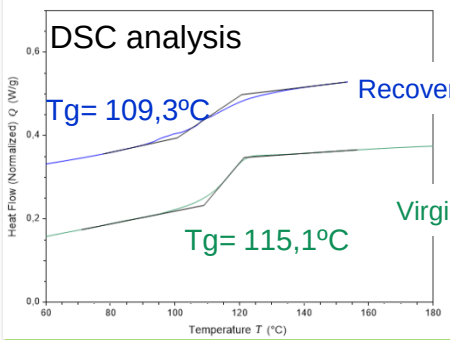
Extraction of the resin by coagulation

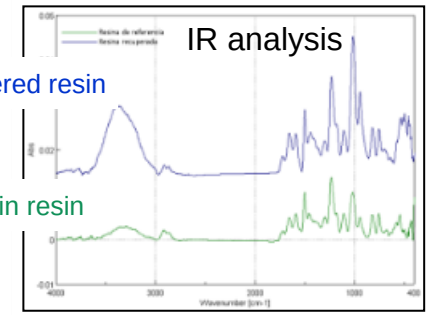


Resin film

Hot compression moulding (1 min @ 200ºC & 5 bar)

Recovered resin (solid after drying)





4. Conclusions

1. The use of dynamic covalent chemistry allows to manufacture a new generation of thermoset composites that preserve their high performance, while showing new unprecedented features once the composite is completely cured, such as Reprocessability, Repairability and Recyclability.

2. Throughout the project, several 3R based materials & processes have been developed with the support of simulation tools to validate the 3R functionalities by manufacturing two demonstrators at pre-industrial scale for automotive and aeronautics. In parallel, the processes to repair the 3R composites as for delaminations and resin microcracks, and the processes to recycle the 3R composites by chemical or mechanical methods have been developed.

3. The important progress made during the project also establishes the bases to continue advancing in the optimization of the 3R concept, in order to adapt it for each specific case and introduce it to the market.

Thank you very much for your attention

Contact details:

nmarkaide@cidetec.es

Tel: +34 943 31 82 12

N. Markaide[a], A. Ruiz de Luzuriaga[a], G. Hoyos[a], A. Rekondo[a], H. Grande[a], S. Fita[b], B. Redondo[b], Fco. José Prado[c], M. Martín-Gallego[d], I. Ruiz de Eguino[e]

[a] CIDETEC, Polymers and Composites Unit, Paseo Miramón 191, E-20014 San Sebastián - Spain
[b] AIMPLAS, Composites Department, Calle Gustave Eiffel 4, 46980 Paterna - Spain
[c] SOFITEC, P. Ind. y Log. de Carmona Manzana LB Nave 1, 41410 Autovía A4, km 521, Sevilla - Spain
[d] GONVARRI STEEL SERVICES, Prolongación de embajadores s/n, E-28053 Madrid - Spain
[e] SEGULA; Avda. Bruselas 8, OF. 8, E-01003 Vitoria-Gasteiz – Spain

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