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Numerical Approach to the Simulation of Vitrimer Matrix Composites Manufacturing by Infusion and Coating by Cold Spray

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Abstract. Aiming to minimize time, energy, and materials-consuming trial and error experimental analyses, a numerical modeling approach of vitrimer flow and cold spray deposition is proposed in this work. The characteristics of vitrimeric matrices were evaluated by elaborating data from previously performed differential scanning calorimetry and dynamic mechanical analysis. The pieces of information related to the transition temperatures and mechanical evolution after curing were exploited to feed the numerical models and to run sensitivity analyses. The flow model is based on prior evaluation of the dry reinforcement permeability at a micro- and meso-scale. The flow model has been implemented using a commercial simulative environment based on the control volumes approach. A single impacting particle was simulated in a finite element environment to analyze, in a focused way, the deposition mechanisms. The objective of this analysis is the integrated implementation of a numerical model for vitrimer flow through carbon fabric reinforcement in infusion processes and single particle deposition on vitrimer matrix composite substrates.

Introduction

In current industry, fiber-reinforced polymers represent a relevant solution to achieve high strength and properties customization coupled with low weight [1]. These features made the composite materials particularly appealing for both high performance and sustainability during the in-service life [2]. The mechanical behavior of these multi-phase materials is based on the capability of precisely orienting the continuous reinforcing filaments [3]. Due to this reason, most processes in high-volume production are evolving towards automation and robotization [4], [5], [6], [7]. In this context, research community is devoting relevant efforts towards the development of composites that are reparable, reformable, weldable, and which can be coated by metal and metallic alloy particles.

Vitrimeric resin systems represent a disruptive innovation in polymers. Vitrimers, a novel class of covalently crosslinked polymers capable of network rearrangement through dynamic covalent exchange reactions, combine the mechanical stability of thermosets with the reprocessability of thermoplastics [8]. Their unique viscoelastic and time-dependent mechanical behavior makes them promising candidates for next-generation functional coatings and hybrid composites. This molecular structure enables them to be reprocessed after curing for repairing or to operate further manufacturing steps. In particular, the authors are studying the feasibility of cold spray depositions on vitrimer matrix composites [9], [10].

Cold spray is a solid-state deposition process in which metallic particles are accelerated to supersonic velocities and impact a substrate to form coatings without melting [11]. Over the past two decades, a substantial body of experimental and numerical research has been devoted to understanding the mechanisms of particle impact, deformation, and bonding in cold spray, particularly in metal-to-metal and metal-to-polymer systems [12]. Despite these advances, most studies have focused on thermoplastic or thermosetting polymer substrates [13], whereas vitrimer materials have received little attention.

To the authors' best knowledge, no previous numerical investigations have addressed the adhesion mechanisms between metallic particles and vitrimer substrates in cold spray conditions.

The present work aims to fill this gap by developing a finite element (FE) model of a single copper particle impacting an epoxy vitrimer substrate. This study represents a first step toward understanding the feasibility and underlying physics of metal-vitrimer deposition via cold spray by numerical approach. This study considers the carbon reinforced vitrimers manufacturing by infusion and the successive coating of their surface by cold spray. In particular, the activity focuses on the numerical modeling of the processes. The numerical approaches, previously implemented and validated on conventional materials, are applied in this study considering an innovative vitrimeric matrix.

Numerical Procedures

Modeling of vitrimer infusion.

The vitrimer considered is an epoxy-based system enriched with zinc acetil-acetate, and it has been widely described and characterized in previous works [14]. The model implemented aims to simulate the infusion of carbon fiber fabrics (twill 2-2, 380 g/m^2, 12k tow) within a square plant mold having dimensions of *100 mm* in length and width and depth of 5 mm. The reinforcement is constituted by 3 layers of carbon fabrics, and it has been concentrated at the lower 3.5 mm of the mold in order to leave a sufficient layer of non-reinforced vitrimer to safely receive the successive cold spray deposition avoiding the erosion of the filaments. Due to this reason, the successive cold spray model can consider a pure vitrimeric substrate, excluding the presence of fibers in the portion of material involved.

The infusion models are based on Darcy's law correlating the vitrimer flow velocity $\bar{u}$, the permeability tensor of the dry reinforcement $\bar{\bar{K}}$, which in this analysis is assumed to be one unique porous medium, the rheological behavior of the impregnating flow η, and the three-dimensional pressure gradient $\bar{\nabla}P$ realized by the vacuum pump into the mold cavity. Darcy's law definition is expressed as:

$$\bar{u} = -\eta^{-1}\bar{\bar{K}}\,\bar{\nabla}P \ . \qquad (1)$$

The evaluation of the permeability is based on the analysis of the average distribution of the fibers in the tow, on their dimensions, concentration and on the geometrical features of the fabrics [15], [16]. Experimental and analytical studies demonstrated that during the infusion, the porosity and the thickness of the fibrous medium undergoes variations due to the absorption of resin by the fabrics [17]. Nevertheless, in the present analysis, this effect has been neglected due to the rigid mold. The fabrics were observed in dry conditions and after impregnation in order to define their geometrical characteristics and the cross-section of the tows. Fig. 1 illustrates the process from geometry acquisition to the digitalization of the fabrics. A dual scale permeability model has been implemented following the procedure described by the scientific literature [18].

The rheology of the vitrimer has been evaluated based on the results of the dynamic mechanical analysis (DMA) reported in a previous publication from the same group [19], by considering in each temperature condition the stress σ^*, the strain ε and the testing frequency ω according to the following model:

$$\eta = \sigma^*/(\varepsilon\,\omega) \ . \qquad (2)$$

The process simulated is a vitrimer infusion in a rigid mold replicating the process described in a previous work [19]. The initial temperature of *75°C* was set for the entire domain. The temperature of *75°C* has been set as a boundary condition for the lower mold, while adiabatic wall condition has been set for the upper mold. The resin is infused at a temperature of *80°C* with a pressure gradient of *0,94 bar*. In conventional infusion processes, the resin preheating is a consolidated practice to improve the reinforcement impregnation and reduce the cycle time [20], [21] The model has been implemented in the numerical environment of the commercial module Ansys CFX.

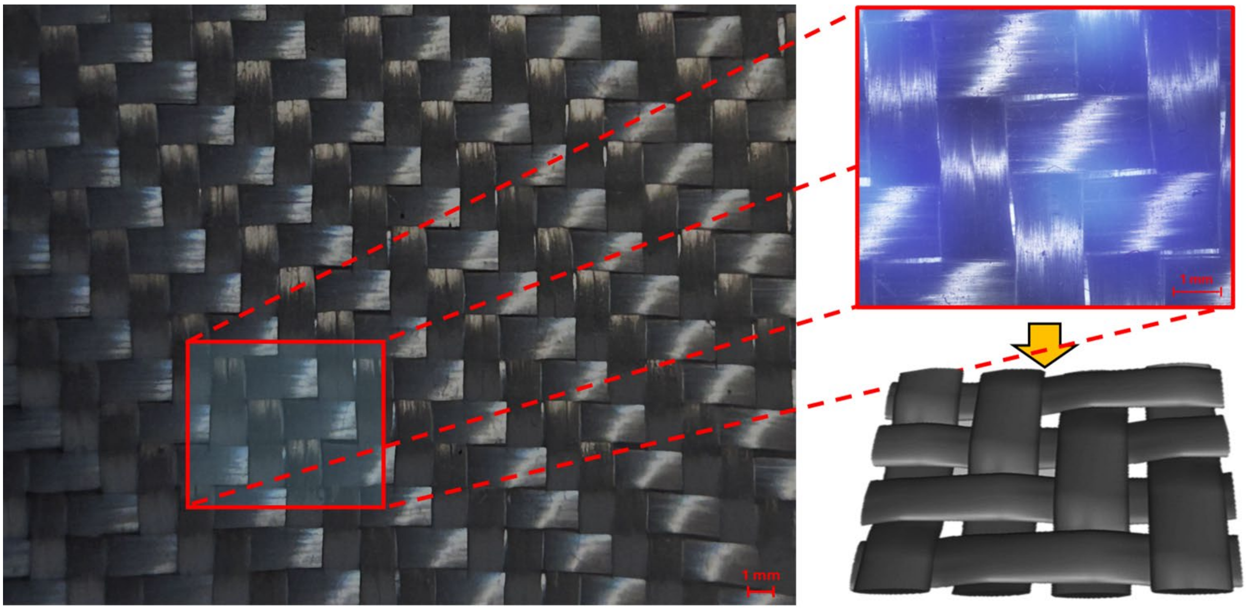

Fig. 1. Observation and digitalization of the reinforcement for permeability prediction.

Modeling of cold spray deposition.

In this section, the authors describe the finite element (FE) modeling of a single copper particle impacting an epoxy vitrimer substrate, conducted using a commercially available FEA software package. Given the novelty of this study, the initial numerical approach assumes the substrate to be unreinforced. The following subsections detail the geometric parameters and material models employed in the simulations.

Both the particle and the substrate were modeled using a Lagrangian reference frame, a common approach in literature for simulating high-velocity impacts while minimizing computational effort [22], [23]. The simulation considered a spherical particle impacting perpendicularly onto a substrate that was at least five times larger than the particle, implemented through a 2D axisymmetric model using explicit dynamic analysis with adiabatic heating effects included. The chosen size ratio ensured that elastic wave reflections from the boundaries did not interfere with the particle-substrate interface during impact [24].

The particle diameter, d_p, was set to 18 µm. The computational domain was divided into several regions to achieve a refined mesh with a gradual transition in element size near the particle-substrate interface. Both the particle and the substrate were discretized using 4-node reduced integration elements (CAX4R). The nominal mesh size for the particle was $d_p/25$, and the same mesh density was applied to the substrate interface region to maintain solution accuracy. This mesh configuration was validated through a mesh sensitivity analysis, which is not included here for brevity. The bottom and right edges of the substrate were fully constrained in all degrees of freedom.

The particle's impact velocity and the initial temperatures of both the particle and the substrate were defined as initial conditions. The effects of gravity and air resistance were neglected. Friction and heat generation due to plastic deformation were incorporated into the model. The tangential contact behavior was governed by a friction coefficient using the *Surface-to-Surface* algorithm, with a value of 0.35 assigned to all contact surfaces [25].

The details of the schematization and the resulting FE model of the impact system are shown in Fig. 2.

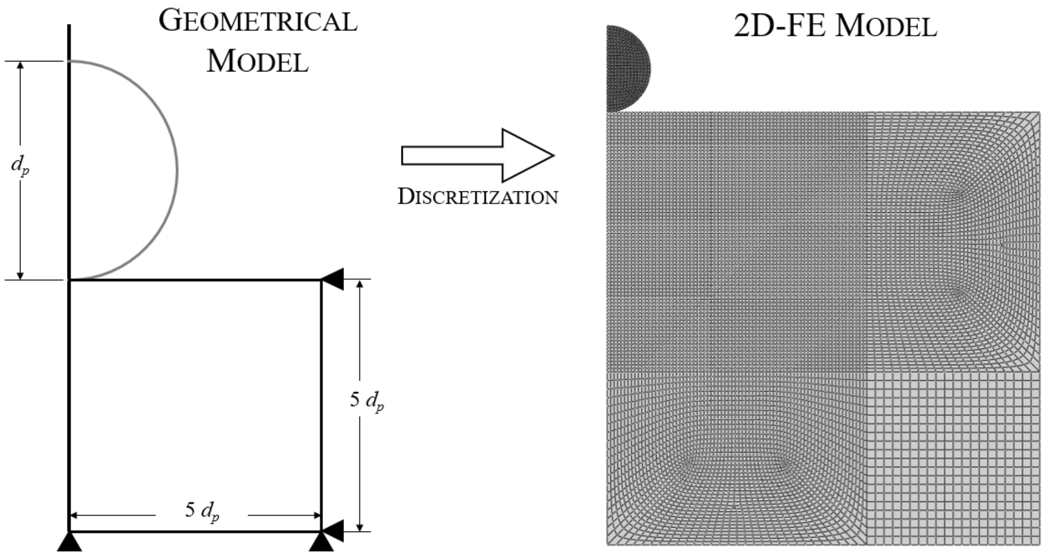

Fig. 2. Schematization (not in scale) and resulting FE model of single particle deposition.

The impact simulations were carried out by varying the velocity of the particle (Vi) between 50 m/s and 400 m/s, with an increasing step of 50 m/s. In order to capture the different deposition mechanisms on vitrimers, the simulations were performed at temperatures below, slightly above, and significantly above the freezing temperature of the epoxy-based vitrimer (Tv=170 °C as found in previous work [26]), namely: T1= 25 °C, T2=175 °C, T3=220 °C. The percentage penetration depth after the impact was calculated as control parameter, according to Eq. 3. Note that H_s and $H_{s,i}$ are the substrate height before and after the impact, respectively.

$$Penetration\ depth\ [\%] = \frac{H_s - H_{s,i}}{H_s} \times 100 \quad . \tag{3}$$

The material modeling of the epoxy vitrimer substrate was performed under the assumption that the material exhibits thermosetting, brittle behavior below the vitrimeric transition temperature and ductile, thermoplastic-like behavior - similar to PMMA polymer - above Tv. Based on this temperature-dependent behavior, the Johnson-Cook (JC) plasticity model was adopted to characterize the mechanical response of both the substrate (above Tv) and the copper particle. The relevant material parameters obtained from the literature are summarized in Table 1 [27], [28], [29].

Table 1. Material parameters for copper and vitrimer.

Material parameter	Copper	Vitrimer	
		Epoxy (T< T_v)	PMMA (T> T_v)
Density, [kg/m^3]	8960	1180	1180
Young's modulus, [GPa]	124	3.6	3.3
Poisson ratio	0.34	0.3	0.35
Tensile strength [MPa]	\\	60	\\
Thermal conductivity, [W/m °C]	386	0.25	0.25
Heat capacity, [J/kg·°C]	383	1470	1470
Melting temperature, [°C]	1083	\\	270
A, [MPa]	90	\\	55
B, [MPa]	292	\\	312
n	0.31	\\	0.62
C	0.025	\\	0.105
Reference strain rate, $\dot{\varepsilon}_0$, [s^{-1}]	1.0	\\	0.001
Thermal exponent, m	1.09	\\	0.8
Inelastic heat fraction	0.9	\\	0.9
Reference temperature, T_R, [°C]	298	\\	170

Results and Discussion

The rheological behavior evaluated from DMA data is reported in Fig. 3.

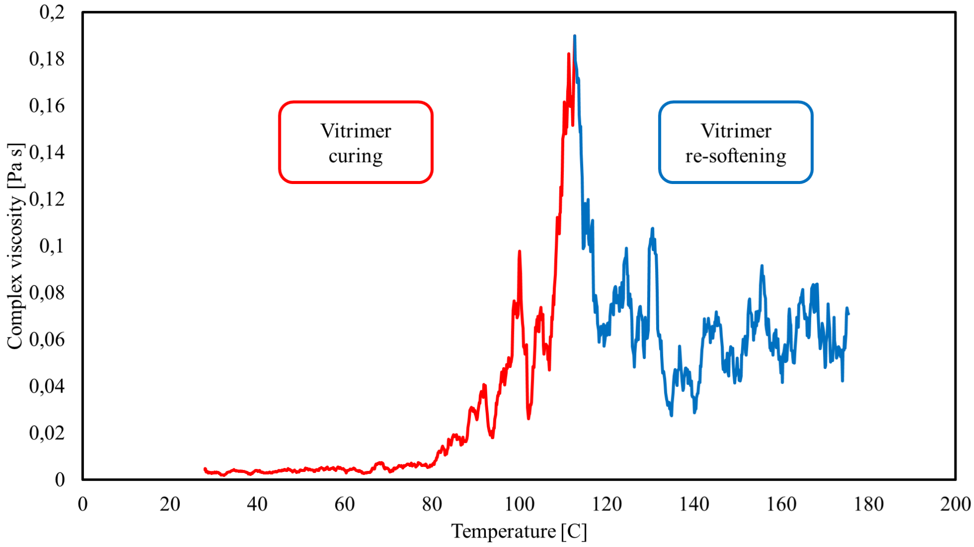

Fig. 3. Rheology of vitrimeric material.

In the specific testing conditions (frequency of *1 Hz* and heating rate of *3°C/min*) the vitrimer curing activation occurs at *79°C* and the viscosity sensibly increases. Once the peak is reached (temperature of *112°C*, viscosity of *0.190 Pa s*) the material exhibits the typical vitrimer resoftening, with a new decrease of viscosity reaching values oscillating around *0.06 Pa s*. The rheological behavior of the considered vitrimeric system can be described by the following model:

$$\eta = \begin{cases} 0.0093\exp[0.082(T - 79°C)]\ ,\ T < 112°C \\ 0.1216\ exp[-0.049(T - 112°C)]\ ,T \geq 112°C \end{cases} . \quad (4)$$

This rheological behavior has been included into Eq. (1) to model the matrix flow through the fibrous preform. The vitrimer infusion simulation evidence that the unsaturated flow crosses the mold length of *100 mm* in *64 s*, and the full saturation of the reinforcement occurred in *240 s*.

This section presents the results of the finite element (FE) modeling. Specifically, when the particle impacts the substrate at 25 °C, the vitrimer exhibits behavior characteristic of a thermosetting, brittle material, as shown in Fig. 4. Particularly, it can be seen that no plastic deformation occurs in either the particle or the substrate, and material damage initiates at an impact velocity of 50 m/s. The von Mises stress distribution and the substrate damaged zone are shown in Fig. 4a and Fig. 4b, respectively, under these conditions.

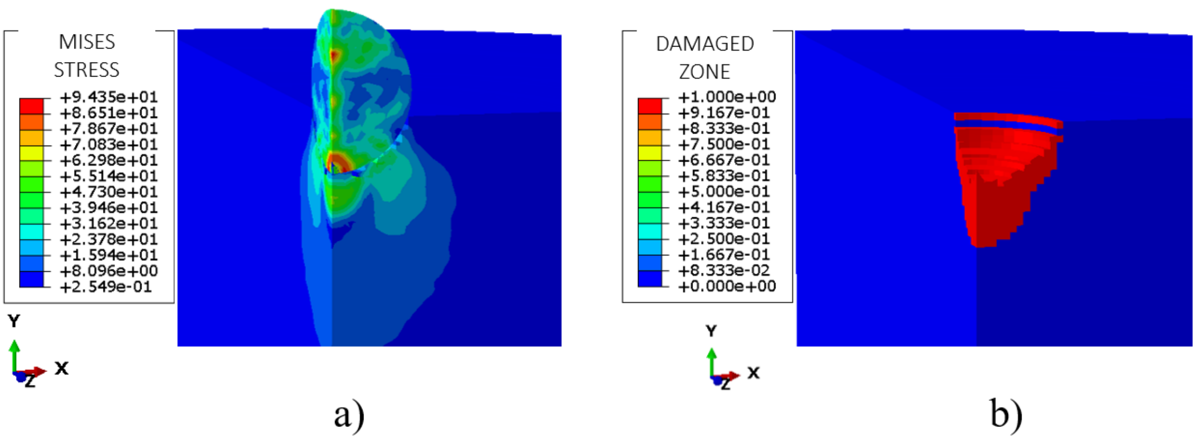

Fig. 4. The von Mises stress distribution (a) and the substrate damaged zone (b) at V_I = 50 m/s and T_I.= 25 °C.

With increasing impact velocity, the extent and severity of polymer damage become more pronounced, thereby inhibiting adhesion between the particle and the substrate, as shown in Fig. 5. Notably, at an impact velocity of 250 m/s, severe damage occurs, and the particle tends to fully penetrate the substrate, disrupting its structure. This behavior is clearly illustrated in Figs. 5a and 5b.

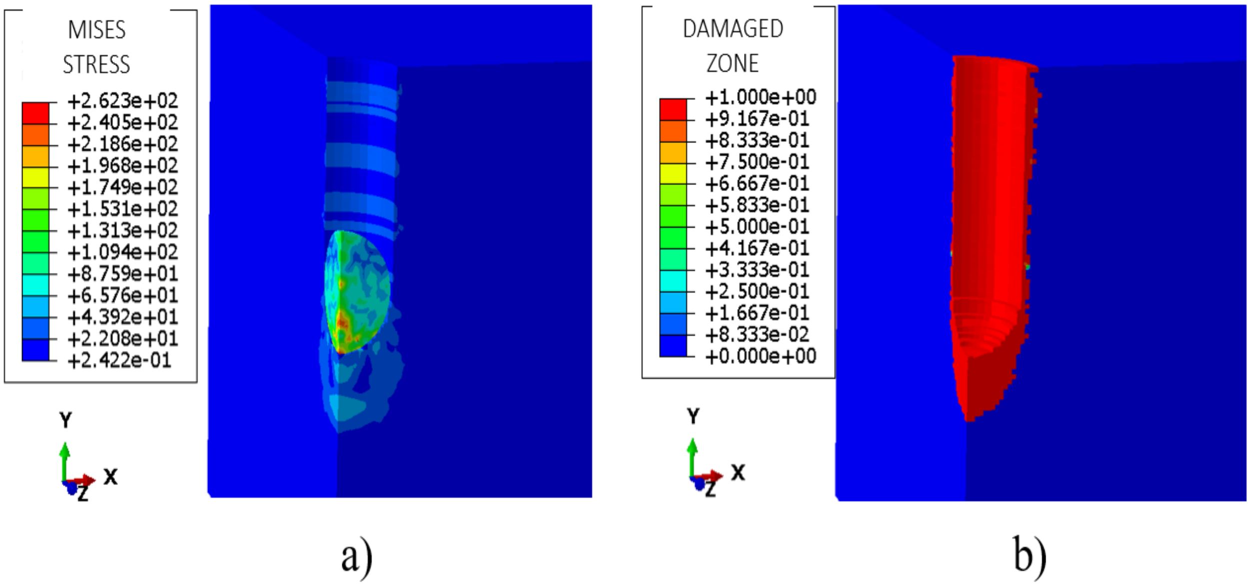

Fig. 5. The von Mises stress distribution (a) and the substrate damaged zone (b) at V_I = 250 m/s and T_I= 25 °C.

When the particle impacts the vitrimer substrate at temperatures exceeding the vitrimer transition temperature, a markedly different behavior is observed compared to impacts occurring below this threshold. At relatively elevated temperatures, the substrate exhibits pronounced ductile characteristics, which facilitate the partial or complete penetration of the impacting particle into the polymeric matrix. As a consequence, the particle becomes embedded within the softened polymer network.

Figure 6 illustrates the influence of the substrate temperature on penetration depth as a function of impact velocity. The plot clearly shows that penetration depth increases monotonically with impact velocity for both thermal conditions. However, for any given velocity, the penetration depth achieved at 220 °C is consistently higher than that at 175 °C, with the difference widening as the velocity increases. This trend indicates that elevated substrate temperatures promote deeper material deformation, likely due to enhanced thermal softening and reduced resistance of the substrate to impact-induced flow. The qualitative interpretation is supported by the von Mises stress contours shown on the right side of the figure.

Overall, the combined trend curves and stress maps confirm that substrate temperature is a dominant parameter governing the impact dynamics. The embedding mechanism is strongly temperature-dependent and becomes increasingly significant as the impact temperature rises well above T_v. Under these conditions, the polymeric substrate undergoes localized deformation, allowing the particle to settle or "accommodate" within its surface. This process enhances the formation of a mechanical interlocking interface between the particle and the substrate. Summarizing, the combined effects of increased ductility, particle penetration, and interfacial interlocking at temperatures above T_v. contribute to a more robust and adhesive particle-substrate interaction.

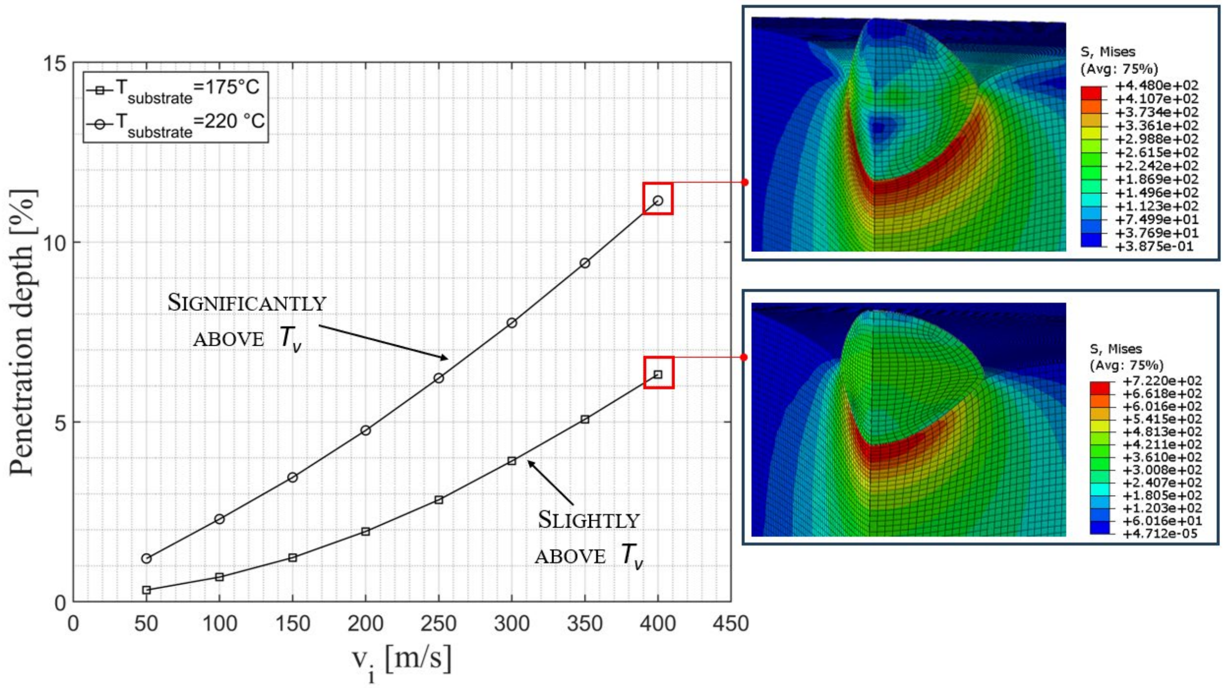

Fig. 6. Penetration depth percentage vs. impact velocity.

Conclusion

A numerical model was successfully implemented to simulate the infusion of vitrimeric resin into carbon fiber reinforcements using Darcy's law and a dual-scale permeability approach. Rheological analysis identified that curing activation occurs at 79 °C, reaching a peak viscosity at 112 °C before the material undergoes a characteristic vitrimer resoftening phase. The simulations showed that the resin flow covers a mold length of 100 mm in 64 seconds, with full reinforcement saturation achieved in 240 seconds.

Finite element (FE) modeling demonstrated that the substrate temperature is the dominant parameter governing impact dynamics and bonding mechanisms. Below the transition temperature the vitrimer behaves as a brittle thermoset; high impact velocities (e.g., 250 m/s) result in severe structural damage and inhibit particle adhesion. Above transition temperature, the substrate exhibits ductile characteristics that allow the copper particles to penetrate and become embedded within the polymer matrix. Higher temperatures (e.g., 220 °C) promote deeper penetration and the formation of a mechanical interlocking interface, leading to more robust and adhesive interaction.

The application of high versatility vitrimer matrices in high performance composites will be based on the exploitation of their unique properties combining good flowability and capability to be resoftened. Future studies should improve the manufacturing processes of the coated vitrimeric composites, investigate their surface properties and explore their reparability potentialities.

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