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Positioning of a self-reinforced polyethylene in the industrial composites market

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Abstract. Self-reinforced composites combine lightness and increased recyclability than conventional composites. To position them within the industrial composites market (conventional composites) and simultaneously highlight their potential in a given application context, it is necessary to analyze their mechanical properties for possible exploitation. For this purpose, compression molding has been used to manufacture different composites. In this short paper, the specific tensile properties of polyethylene matrix composites reinforced with glass, carbon, flax, and polyethylene fabrics, respectively, are compared at room temperature. The results give an excellent perspective to self-reinforced polyethylene.

Keywords: compression molding / reinforcement / mechanical properties / self-reinforced polyethylene

Résumé. Positionnement d'un polyéthylène auto-renforcé sur le marché des composites industriels. Les composites auto-renforcés allient légèreté et recyclabilité accrue par rapport aux composites conventionnels. Pour les positionner au sein du marché des composites industriels (composites conventionnels) et simultanément mettre en évidence leur potentiel dans un contexte applicatif donné, il est nécessaire d'analyser leurs propriétés mécaniques en vue d'une éventuelle exploitation. À cette fin, le moulage par compression a été utilisé pour fabriquer différents composites. Dans ce court article, les propriétés spécifiques de traction de composites à matrice polyéthylène, renforcés par des tissus de verre, de carbone, de lin et de polyéthylène, respectivement, sont comparées à température ambiante. Les résultats donnent une excellente perspective pour l'exploitation du polyéthylène auto-renforcé.

Mots clés : moulage par compression / renforcement / propriétés mécaniques / polyéthylène auto-renforcé

1 Introduction

In the context of the current energy transition, reducing energy consumption is a significant issue, especially in the field of transportation and, in particular, in the automotive and aerospace industries. The use of composites in such a field allows for lightening the structures by replacing denser metallic structural parts and thus reducing in part the energy needed to move. It is then a question of taking advantage of the benefits of different materials and, in particular, of stiffening certain polymers with specific high stiffness fibers to extend their range of use. These

composites, for example, with an organic matrix and carbon or glass reinforcements, have been the subject of much work over the last forty years. Their behavior is now perfectly mastered, and these materials have found their place in the market, and they represent a large part of this one. More precisely, in 2018, glass fiber reinforced composites represented 95% of the composites present on the market [1].

However, a new major ecological issue is emerging today and is pushing us to reconsider the carbon footprint & energy consumption in the entire life cycle of the parts produced. Indeed, the recycling of common polymers is well mastered. Still, the recycling of mixtures, as in the particular case of composites, is more complicated and can lead to a separation step of the species, a costly action.

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Also, when the nature of the fibers is different from that of the matrix, recycling may involve the use of complex chemistry or energy-intensive processes. For example, for a carbon-reinforced composite, pyrolysis in an inert atmosphere reactor and depolymerization have been proposed to reuse the materials [2]. To overcome this problem, a concept of composites has been proposed by Capiati and Porter [3]. This is the self-reinforced polymer (SRP) concept, where the reinforcement and matrix are similar or even of identical chemical natures. Therefore, strong interactions can be expected between matrix and reinforcement.

Because of this meso-structure using a single polymer in different phases, SRP has higher recyclability. Therefore, recycling is intrinsically favored, and it is thus possible to remelt or grind and then extrude these materials to recycle them [4,5]. In addition to the conditions favorable to recycling a SRP, this type of composite combines the ease of recycling of thermoplastic polymers, whose nature is based on the weak bonds between the macromolecules for the families of polymers considered. Concerning SRPs, whether they are in the form of SRPET [6] (Self-Reinforced PolyEthylene Terephthalate), SRPP [5,7] (Self-Reinforced PolyPropylene), or SRPE [8] (Self-Reinforced PolyEthylene), it has been shown that they seem to be good candidates for upcycling. This property promotes interest in SRP materials.

Although their recyclability is a fundamental driver for their development, the only commercialized SRP today are SRPP. (Self-Reinforced PolyPropylene) [6,9]. Indeed, the adoption of these materials of interest is relatively slow due to a lack of knowledge associated with how to process them, their complex behavior, and their reprocessing, reasons for which a general caution in the choice of new technology must be added [10].

Moreover, to consider replacing more conventional composites for certain structural parts, the assurance of good mechanical resistance is also necessary. Some comparative studies have been conducted in the literature. It has been pointed out that regardless of the nature of the SRP, the mechanical properties of SRPs fall between those of the associated isotropic polymer and the glass fiber reinforced polymer [9]. While the tensile strength and modulus of glass fibers are much higher than those of, e.g., PP (PolyPropylene) fibers, the specific modulus, and strength of SRPP can, in contrast, compete with those of glass fibers [4]. This is justified by the close chemical nature of the different constituents. Thus, this can lead to a direct and optimal cohesion at the interface [3,11,12].

Thus, the interface seems to be a particular issue. A study of the nature of this one was done by Ajji et al. [13] for Ultra-High Molecular Weight PolyEthylene (UHMWPE) – (Spectra[®] 1000 fabric)/LDPE (Low-Density PolyEthylene), Kevlar/LDPE, and glass/LDPE composites. The efficiency of UHMWPE fibers was found to be comparable to Kevlar fibers and better than glass fibers. This was justified by the unique nature of the bonding between the fibers and the transcrystalline region for UHMWPE/LDPE composites. In addition, the longitudinal properties of a SRUHMWPE (Self-Reinforced) composite showed

excellent properties when compared to a UHMWPE/epoxy or Kevlar/epoxy, or UHMWPE/HDPE (High-Density PE) composite [14].

Finally, the impact properties of SRPs were also compared to those of more conventional composites. Thermoplastic fibers, which are more ductile than carbon or glass fibers, allow for the absorption of a greater amount of energy since they can plastically deform [15]. In addition, the region between the fibers is the one that is capable of absorbing energy [16]. Improved toughness, compared to glass or natural fiber reinforced iso-matrix composites, has also been reported for SRP [4,9,15,16].

In the literature, the comparison with more conventional composites is generally made with a PP because the latter presents a combination of high toughness, good thermal properties, and low cost. In all these studies, specific parameters of their behavior are compared, and it appears quite clearly that SRP can be a choice when designing structural parts with more or less load. However, to our knowledge, the comparison of the potential of SRPEs has not been addressed in the literature. Nevertheless, due to the remarkable properties of UHMWPE fibers [17], it would be interesting to evaluate them by imposing the same reinforcement mass, for example.

Thus, this short article aims to position SRPE among the most commonly used composites in the industry. To compare the behavior of the different composites of this study, several choices are possible: either to compare the behavior of specimens made at an iso-volume fraction of reinforcements or specimens with identical weights and volumes. The targeted applications are rather in the field of transport, for which the weight constraint is significant. It then appeared to us opportune (but debatable) to choose a comparison with identical weight. A key point is then the ratio between the stiffness and the weight of the sample. As the specimens have a similar volume and weight, the stiffnesses can be compared with each other, and they correspond to so-called specific quantities.

Thus, several composites were processed in an identical way and at iso-matrix, and their specific behavior was compared. This SRPE has already been extensively characterized in tensile [17] and creep [18], a characterization that highlighted the value of such a composite. The results presented in the following will show the interest of this material for some industrial applications.

2 Material and methods

2.1 Materials

Different composites have been designed with the same thermoplastic matrix. It is a metallocene (mPE) based on a Low-Density PolyEthylene (LDPE), produced by Total-Energies One Tech and available in granules. The melting temperature indicated on the data sheet is 108.5°C. Further technical data was provided in previous work [19]. The reference for this grade is confidential.

Four composites were manufactured with different natures of reinforcement (glass, carbon, flax, UHMWPE) in commercially available fabrics, whose properties are shown in Table 1. The architecture of the glass, carbon and

Table 1. Properties of the different industrial fabrics used.**Table 1.** Propriétés des différents tissus industriels utilisés.

Properties	Fabrics			
	Glass	Carbon	Flax (40% Flax/PP)	UHMWPE (Spectra 1000 [®])
Supplier	Sandtech	Goodfellow	Goodfellow	Goodfellow
Weight (gr/mm ²)	200	200	420	88
Thickness (mm)	0.3	0.3	0.84	0.17
Type of fabric	Plain	Plain	Twill 2 × 2	Plain

Table 2. Details of the composition of the study materials.**Table 2.** Détails de la composition des matériaux de l'étude.

Composites	Nature of the matrix	Nature of the reinforcing fabrics	Number of reinforcement layers introduced	Number of matrix films and thickness	Volume fraction of reinforcement in the composite plate
SRPE	mPE	UHMWPE	4	5 films of 1 mm	0.14
PE/glass	mPE	Glass	2	3 films of 1.7 mm	0.12
PE/carbon	mPE	Carbon	2	3 films of 1.7 mm	0.15
PE/flax	mPE	Flax	1	2 films of 2.5 mm	0.18

UHMWPE fabrics is identical; it is plain fabric and therefore perfectly balanced. This choice was made to minimize the effect of the fabric architecture. However, in the case of flax, the fabric is a 2 × 2 twill.

However, the process conditions are similar in each case to ensure an iso-transformation of the matrix. Therefore, the comparison between composites reinforced with carbon, glass, flax, and UHMWPE fabrics is made at iso-weight of reinforcements. The tested specimens have the same dimensions and thickness, so the same volume and total mass, including 2.6 g of reinforcement introduced into a composite plate. The composition of the different composites of the study is detailed in Table 2. The volume fraction of reinforcement indicated corresponds to that introduced into the composite plate before extraction of the specimens to be tested, 4 per composite plate. Therefore, the different volume fractions are of the same order.

2.2 Compression molding process

A compression molding process was used on an Instron 4505 tensile machine, using an inverter system, heating platens, and a mold and a fabric holding system. This process was presented in previous work [19].

The process is done in two steps and is similar to a film stacking process [20]. First, PE (PolyEthylene) films, thin sheets, are manufactured according to the protocol [19] with the desired thickness. Then, the films and fabric pieces are stacked in a second step, with the fabrics always between two polymer films. The fabrics are held with the device and are not tensioned. Then, the temperature-pressure cycle is applied to obtain the final composite plate. The different composites are all processed at 140°C and 10 kN during the same test time. Before demolding the plates, the mold is cooled to room temperature. The thickness of each of the final plates is similar.

2.3 Tensile characterization

The tensile tests were carried out on an INSTRON 1195 machine with screw jaws and a 5 kN load cell. In the form of dumbbells, the specimens measure 60 × 15 × 4 mm, and the useful area is 10 × 6 × 4 mm. The shape is similar to a creep behavior study sample conducted on a high-density polyethylene [21]. These were machined within the compression-molded plates. The tests were conducted at room temperature on at least four specimens for each composite type, extracted from two plates made for the different conditions.

The machine is driven at a constant strain rate of 6%/min. This control is possible thanks to tracking four markers, two in the longitudinal direction (tensile direction) and two in the transverse direction, using Videotraction[®]. This is an optical technique developed by G'Sell et al. [22]. An acquisition is made every 100 m/s. The calculation of the true stress at each acquisition step is established under the assumption of transverse isotropy. In addition, this method considers the non-homogeneous reduction of cross-sections in tensile and creep tests [21].

The initial modulus was calculated in the strain range of 0.05 to 0.25% for the different composites and between 0.05 and 0.5% for the unreinforced PE matrix.

2.4 Tomographic analysis

One non-destructive technique to determine the manufacturing quality of composites in the sample volume is to use tomography. This technique allows, among other things, to reconstruct a 3D image of the sample from 2D images.

An UltraTom tomograph from RX Solutions was used with the following characteristics: a selected source of 160 kV and a resolution of approximately 6 μm/pixel

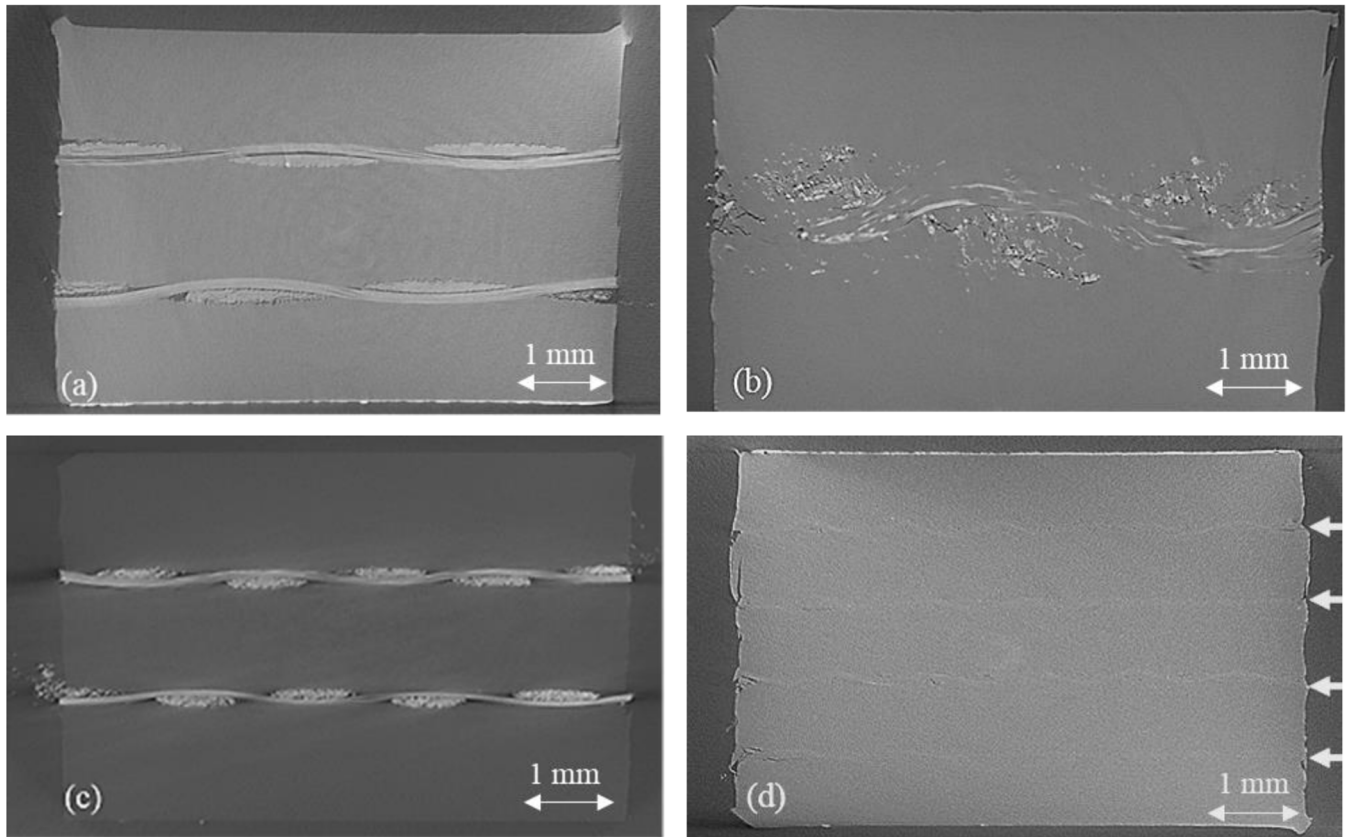


Fig. 1. Tomographic images of PE matrix composites reinforced with (a) carbon, (b) flax, (c) glass, and (d) UHMWPE fabrics.

Fig. 1. Images tomographiques de composites à matrice PE renforcés par des tissus (a) de carbone, (b) de lin, (c) de verre et (d) d'UHMWPE.

obtained using a voltage of 60 kV and a current in the tube of 100 μ A. Furthermore, to perform tomography of the whole specimen, a helical type of acquisition was chosen.

3 Results and discussion

3.1 Validation of the quality of the composites

Initially, a tomographic analysis was conducted to verify the proper impregnation of the matrix within the reinforcing fabrics. Because the densities of the fabrics are different and the plates of the various composites were manufactured with iso-weight of reinforcements, the number of layers embedded in the composites studied differed, and so did the number of films separating them. An image of the thickness of a sample of each of the four types of composites is shown in Figure 1. The entire thickness of the samples is observable. For each of the composites in the study, the impregnation of the matrix within the fabrics appears to be satisfactory. Given the quality of the manufactured composites, it can be asserted that the behavior highlighted is well representative of the behavior of the corresponding composite.

The contrast that can be observed on the tomograph is dependent on the density of the material. In Figures 1a and 1c, the plain fabric is well balanced. For the SRPE, in

Figure 1d, UHMWPE fabric/PE matrix, the contrast between the two constituents is less critical than the other composites observed. Although this composite consists of a single material with identical atomic numbers, it is possible to distinguish between the reinforcement and the matrix, as the distribution and density of the microstructures differ somewhat. This is directly attributable to the drawing process undergone by the UHMWPE reinforcements. However, the grey shades of the matrix and the reinforcement remain very close. Also, to distinguish the four plies that have been integrated into the composite, white arrows locate them in Figure 1d. The number of reinforcement layers introduced in other composites is identifiable in the tomographic pictures.

3.2 Comparison of the specific properties of the composites used

The specimens obtained were tested in uniaxial tension at room temperature. Therefore, to facilitate the reader's analysis, only one representative curve is plotted for each composite type. These true stress/true strain curves determined are described in Figure 2a.

The shapes of the curves are different depending on the composite being examined:

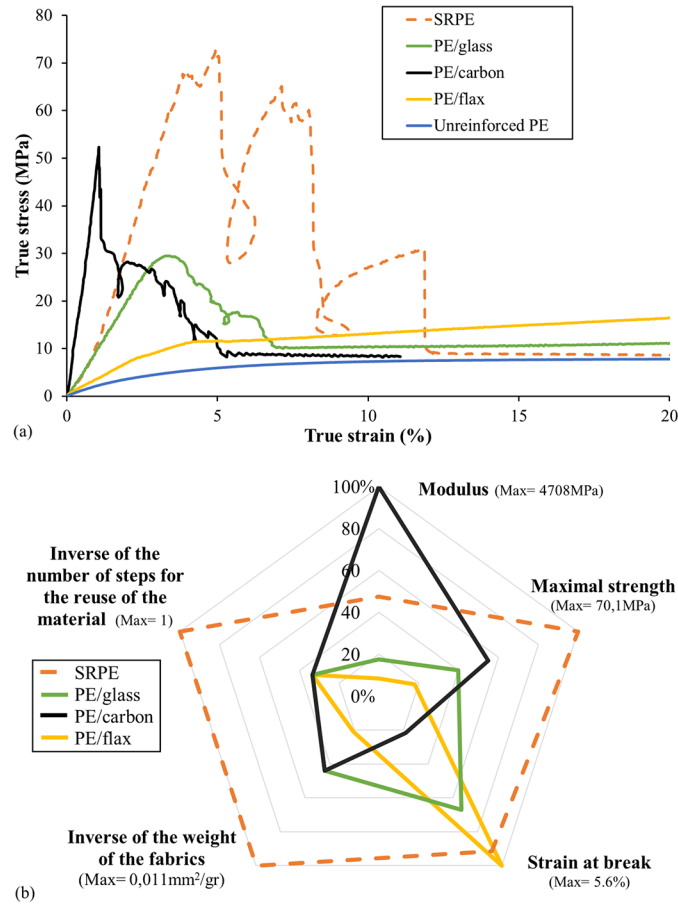


Fig. 2. (a) True stress/true strain curves of iso-matrix composites reinforced with different types of fabrics and tested in uniaxial tension; (b) Radar diagram of the specific mechanical properties of the other composites analyzed.

Fig. 2. (a) Courbes contrainte vraie/déformation vraie de composites à iso-matrice renforcés avec différents types de tissus et testés en tension uniaxiale; (b) Diagramme radar des propriétés mécaniques spécifiques des autres composites analysés.

- In the case of carbon, glass, and SRPE, the behavior is quasi-linear until the rupture of the fabrics. For a small proportion of reinforcement introduced, whatever the nature of the reinforcement, the modulus appears higher than for unreinforced PE.
- For SRPE, the rupture of the different layers of fabrics can be subsequent and show the stress transfer between layers.
- For all the composites, except for the flax reinforcement, the curve appears similar after the subsequent and complete ruptures of the reinforcements. It is superimposed on that of the matrix alone, i.e., the behavior of an unreinforced PE (blue curve in Fig. 2a).
- For flax, a yield point and hardening are noticeable. This may be associated with the architecture of the fabric, which is not plain but twill. This last one is therefore not balanced. The four curves obtained for this composite all showed a similar shape. The tests could be conducted up to an average true strain of 40% in the tensile direction, the end of the test corresponding to a loss of the markers, and thus the stop of the piloting, loss due to the appearance of a necking. In addition, the 2 × 2 twill fabric is less wavy than a plain fabric and therefore has a higher

stiffness. This is a more advantageous case for the flax-reinforced composite. However, it has lower specific properties than all other composites considered.

- In addition, the curve of SRPE is almost identical to that of the glass fabric reinforced composite and then becomes superior to it. An inflection point attributed to the intrinsic nature of UHMWPE reinforcements is noticeable. Previous work has already highlighted its presence and proposed a beginning explanation of its origin [19], but future work will focus more on this point.

Since the composites have an almost identical mass of reinforcement, the specific properties of the different composites could be compared. The specific modulus, taken at the maximum slope for each of the composites before reinforcement breakage, the specific maximum strength achieved, and the specific strain at break of the fabrics are analyzed and represented on a radar diagram in Figure 2b. The strain at break chosen corresponds to the maximum strength in each of the case examined. The data presented are the average data reported to the maximum of each. The ratio is given in percent. The maximums are shown in Figure 2b.

On the other hand, the flax reinforced composite shows a specific maximum strength and modulus well below those of other composites. The type of fabric may also play a role. Moreover, as expected, the carbon fabric reinforced composite shows a remarkable specific modulus. However, SRPE has the most significant specific strength. For the latter, the specific modulus and strain at break are largely comparable to those of the glass fabric reinforced composite and even dominate them (taking into account the maximum modulus after the inflection point). If we disregard the flax-reinforced composite with twill fabric, SRPE is the most ductile composite.

The inverse of the weight of the fabrics is also presented, showing the attractive lightness of a UHMWPE reinforcement. In addition, the ease of material reuse is quantified as a function of the inverse of the number of steps required to separate and reuse the materials, particularly the fibers, that have a high stiffness and can offer an interesting second life. Three steps are generally necessary for composites reinforced with flax, carbon, and glass fibers. Indeed, pyrolysis and depolymerization are necessary to recover the fibers. Depolymerization is often essential to remove any remaining adhesion, especially at the interface, as most fibers must be sized to ensure good interface quality. However, pyrolysis is harmful to the reinforcements, particularly those of glass and flax, because they are somewhat sensitive to high temperatures. In addition, the properties of flax fibers drop with thermal aging. On the other hand, continuous carbon reinforcements, which are very expensive, can be recovered satisfactorily with this method [2].

In addition, direct mechanical grinding of parts made of glass and flax reinforced composites has also been conducted. However, the material with short glass fibers often has inferior properties to the virgin material and is then introduced into matrices rather than reused as such. For natural fibers, high shear rates can lead to significant losses in mechanical performance [23]. SRPE, on the other hand, could be reused as such after a simple mechanical grinding. Due to the similar chemical nature of the materials, no separation of the constituents is necessary. In the radar diagram, the area of the pentagon, corresponding to the specific properties of SRPE, the lightness, and the ease to reuse, is the largest compared to the other composites, which shows its potential.

Besides, the number of PE grades on the market is huge. The grades are classified into prominent families, including LDPE, MDPE (Medium-Density PE), and HDPE, which multiplies the combination possibilities for SRPE design. In addition, composites made from natural fibers have widely varying properties due to the nature of the fibers.

For an equivalent process, SRPE shows specific mechanical properties that are competitive with, for example, an iso-matrix composite reinforced with glass fabrics. However, it can currently have a higher cost, depending on the choice of reinforcements, due to the more complex process that must be applied than that for glass or carbon fibers, but also due to its lower presence on the

market, compared to the last-mentioned fibers. On the other hand, the glass- and carbon-reinforced composites market is more mature, so the costs are relatively optimal.

The market for self-reinforced composites is more confidential, but we could move towards the price of a glass-reinforced composite by increasing volumes. If the cost calculation includes the recycling phase, the financial interest should be there. Moreover, the self-reinforcing character of PE enables us to imagine many more technical applications for PE. It is possible to envisage an application market close to glass fiber reinforced composites while taking advantage of a simply recyclable composite.

4 Conclusion

The use of self-reinforced polymers in some areas seems attractive due to their increased recyclability. However, they must be examined in terms of mechanical resistance to meet the mechanical requirements given in the specifications of the structural part considered. Therefore, to position a SRPE on the composites market, the specific mechanical properties of a SRPE (PE matrix/UHMWPE fabrics) were compared to those of composites reinforced with glass, carbon, and flax fabrics with identical matrices and processed under similar conditions. Even with a low reinforcement ratio and for certain specification conditions at room temperature, these SRPE composites seem to show a promising future due to the specific characteristics determined in tension compared to a glass-reinforced composite.

Conflict of interest

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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Credit author statement

Coline Roiron: Methodology, Validation, Investigation, Writing-original Draft, Writing-Review and Editing, Visualization.

Eric Lainé: Conceptualization, Writing-Review and Editing, Supervision.

Jean-Claude Grandidier: Conceptualization, Writing-Review and Editing, Supervision.

Nicolas Garois: Conceptualization, Project administration, Supervision.

Baptiste Voillequin: Conceptualization, Project administration, Supervision.

Cathie Vix-Guterl: Project administration.

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