The effect of a coupling agent on the impact behaviour of flax fibre composites

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Abstract

The effects of a coupling agent on the behaviour of flax fibre reinforced composites have been investigated by testing the specimens under both quasi-static indentation and high velocity impact loading. The specimens are manufactured embedding a commercial flax fibre fabric in a polypropylene (PP) matrix, neat and pre-modified with a maleic anhydride grafted PP, the latter acting as a coupling agent to enhance the interfacial adhesion. Quasi-static (QS) compressive tests were performed using a dynamometer testing machine equipped with a high-density polyethylene indenter having the same geometry of the projectile employed in the impact tests. The impact tests were conducted setting three different impact velocities. Digital Image Correlation maps of out-of-plane displacement were employed to compare the specimens with and without the coupling agent. The QS testing results indicate that the coupling agent has an enhancing influence on the bending stiffness of tested flax composites. The testing results show that the coupling agent improves the mechanical behaviour by decreasing the out-of-plane displacement under impact loading. This approach gives rise to new materials potentially useful for applications where impact performance is desired whilst also providing an opportunity for the incorporation of natural fibres to produce a lightweight composite

1.Introduction

The interest is growing in industrial and academic circles in the study and development of new eco-friendly materials to replace conventional existing ones. With this trend, natural fibers are identified as an attractive alternative as reinforcements for polymer composites especially due to its lightness, renewability, mechanical properties and low cost (Shanks 2013,[1](Kaboglu et al. 2017)(Kaboglu et al. 2017)(Kaboglu et al. 2017)(Kaboglu et al. 2017)[2]. However, use is still limited by intrinsic drawbacks such as low thermal stability, dependence of the properties on source, extraction techniques and the hydrophilic nature that can lead to undesirable swelling and maceration phenomena. These drawbacks limit its use mainly to the fabrication of non-structural indoor components [3], [4].

Amongst natural fibers, flax has recently been widely considered for many applications in various fields e.g. automotive, building construction, furniture and related applications, for which it has appropriate strength and stiffness [5]. Flax fibers are mainly constituted by cellulose, hemi-cellulose and lignin with minor components such as waxes, other inorganic materials, and bound water.

Polypropylene (PP), a thermoplastic, is widely utilized in many industrial fields such as packaging and automotive. Recently, many composites based on this matrix and reinforced with natural fibers have been investigated to identify new eco-friendly solutions that can be used in at least part of the many industrial applications already validated [6], [7].

Recognizing that one of the most important parameters is the fiber-matrix interface on the mechanical properties [8], [9], special attention has been given to developing the interfacial bond between hydrophobic PP and hydrophilic flax fibers. Although it is expected that the roughness of natural fiber surface will provide a positive contribution on the bonding strength of related composites, several research efforts have focused, for example, to adjust the cellulosic fiber surface with the aim to improve the bond with the hosting matrix including chemical modifications by adding coupling agents [10]–[12].

Bledzki et al. [10] modified flax fibre with acetylation. With the addition of the maleated polypropylene coupling agent, flexural and tensile strength properties increased between 20 to 35%.

Van Den Oever et al. (2000) considered the effect of the physical structure of flax fibres on the mechanical behaviour of flax composites under tensile and flexural loading. This study showed that structural features of fibres have no effect on the tensile modulus of composites that are reinforced and did not ameliorate the interfacial adhesion at the interface with the surrounding matrix. On the contrary, flexural performances were significantly affected by the physical structure of included flax fibres. It was suggested that the strength of flax fibre reinforced PP composites could be increased through further isolation of the elementary flax fibres.

Van de Velde and Kiekens (2003) investigated the effect of the processing of flax fibres on the physical properties of the related composites. They used varying flax/PP processing including retted one needled, retted twice needled, boiled and bleached. They found that boiled flax/PP gave best results compared to the others with respect to mechanical properties.

The same authors also studied the properties of the flax fibre reinforced composites with 12 thermoplastic polymers [13]. They concluded that PP was the best option for protection against water, energy savings in production and application in terms of weight. Furthermore, Ngo et al. [14] investigated the influence of different thermoplastic polymers on the mechanical properties of flax composites. They found that PP was the best option for manufacturing and application.

Cantero et al. (2003) studied the effect of the treatment of the fibre on the mechanical behaviour of the flax composites under three-point bending and tensile loading. The applied treatments were maleic anhydride (MA), MA-polypropylene copolymer (MAPP) and vinyl trimethoxy silane. They showed that the treatment of flax fibres influences flexural and tensile strength of the composites in a desirous way. The MAPP-treated composite produced the best results.

Weyenberg et al. [15], [16] investigated the influence of processing and chemical treatments of flax fibres on the composites by applying tensile and three point bending loading on the specimens. They found that alkalisation involving treatment in a 4% NaOH solution increased the transverse strength as much as 30%. Wang et al. [17] also studied the effect of chemical modifications (mercerization, benzoylation and peroxide treatment) on the tensile and dome forming behaviour of flax composites. The specimens under different conditions (untreated, saturated and dried) were tested under tensile loading. They found that saturated specimens have higher elongation at break and strength compared to the other specimens in untreated and dried conditions. The absorption of water of the chemically treated specimens was lower than that of the untreated fiber-based composites.

Gu and Liyan [18] analyzed the effect of the fabric specification in terms of weft and warp counts on the flax composite properties. They tested three different weft counts (70, 85 and 100) by keeping the same warp count and two different weave structures (plain and twill) under tensile loading. They concluded that the plain weave weft density reduces the tensile strength, however it increases the elongation at break warp. It showed the weave structure having a significant part in the final product tensile strength.

John and Anandjiwala [19] studied the outcome of a zein coupling agent treatment on flax composite by using tensile, three-point bending, Charpy impact and dynamic mechanical tests. They indicated that this chemical pre-treatment of fibres improves the mechanical properties (tensile and flexural strength and modulus) of flax-reinforced composites.

Bella et al. (2010) analysed the mechanical properties of composites with bidirectional flax fabrics having different areal weight and subjected to varying chemical treatments to investigate the effect on the mechanical properties of flax composites. The tests results highlighted tensile and flexural benefits.

Barkoula et al. (2010) performed tensile and impact tests on various flax composite specimens to understand the effect of the compounding process. They found that the inclusion of a maleic anhydride coupling agent to the PP improved composite tensile properties. In addition, long-fibre thermoplastics increased composite toughness. They identified that the use of a kneader represents the best compounding process to obtain flax composites with improved mechanical performances.

Yu et al. (2013) studied the tensile properties of composites including neat and varying chemically treated flax fibres (alkali, MAPP copolymer, and vinyltrimethoxysilane). They concluded that the treated reinforcing fibres improved both the interfacial strength and tensile performance of the investigated flax composites.

El-Sabbagh (2014) investigated mechanical and thermal behaviour effects of natural fibre/PP composites including MAPP. Differential scanning calorimetry (DSC) and thermal gravimetric analysis (TGA) and tests were applied to composites with different coupling agent ratios (0%, 6.7%, 10.0%, 13.3%, 16.7%). Results showed that a content of coupling agent equal to 6.7% is enough for an optimal improvement in mechanical properties of the investigated composite systems. Rahman et al. [20] investigated the influence of fibre orientation and volume fraction on the impact behaviour of the flax composites applying Charpy and drop weight impact tests.

They concluded that the best fibre orientation is 0° under Charpy test and 30° under out-of-plane impact. In addition, 0.31 and 0.40 volume fractions were considered to be good for practical applications in case of out-of-plane impact loading.

Similar to impact studies done on other common composites such as those with the reinforcement phase being carbon-fibre based (H Liu et al. 2019, H Liu et al. 2020a, H Liu et al. 2020b), impact studies on composites based on flax fibre reinforcement are required to sufficiently address the impact performance/properties of these materials. The importance of impact performance is increasing in aviation (such as drones for package delivery) and in automotive applications where lighter frames that can withstand impact are highly desired. This research was therefore devoted to studying the coupling agent effects on the behaviour of flax composites subjected to Quasi-static (QS) compression and high velocity impact loadings at an in-house mechanical testing rig. Digital Image Correlation (DIC) tests were performed to estimate the extent of out-of-plane deformations occurring in the two examined samples.

2. Experimental

2.1 Materials

The composite plates used were based on a polypropylene (PP) matrix (Hyosung Topilene PP J640, MFI@230 °C, 2.16 kg: 10 g/10 min) provided by Songhan Plastic Technology Co. Ltd. and a 2x2 twill flax fabric supplied by Biotex (weight: 200 g/m^2) shown in Figure 1-a. The coupling agent Chemtura Polybond 3000 (MFI@190 °C, 2.16 kg: 400 g/10 min) was applied to the matrix at 2 wt%. Polybond 3000 is a PP grafted with MA (PP-g-MA) with the MA content being typically in the range of 1.1% to 1.3%.

The modification of the commercial PP was performed with the aid of a twin-screw extruder Collin Teachline ZK25T (Ebersberg – Germany), operating from the hopper to the die with a 180-190-205-195-185 °C temperature profile with a 60 rpm screw speed. The neat PP and the extruded pellets of modified PP (PPC) were transformed into flat films with thickness approximately equal to 35-40 μm using a Collin Teach-Line E20T flat die extruder equipped with a calendar CR72T (Ebersberg – Germany). The filming was done at a screw speed of 55 rpm, with a 180-190-200-190-185 °C screw temperature profile. For reinforcement, the flax fabric was used after drying in a vacuum convection oven at 85 °C for 3 hours to reduce its humidity content. These two types of composites were named as Flax/PP which contained neat PP and Flax/PPC which contained pre-modified PP.

2.2 Laminates preparation

Composite laminates were obtained through alternating layers of neat PP or modified PPC films and woven flax fiber fabric by the film-stacking technique followed by the hot-compression of the lay-up using a lab press Collin P400E according to a pre-optimized molding cycle (Figure 2).

Using this technique, square 320 mm x 320 mm plaques consisting of 12 and 30 balanced fabric layers, symmetrically arranged on the laminate middle plane, were produced with a thickness of 4 mm and 10 mm respectively. In both cases, the volume percentage of fiber was 46-47%.

Plates, opportunely trimmed at the edges, were cut into square specimens (140 mm x 140 mm) and drilled at the edge according to the scheme reported in Figure 3 and clamped as shown in Figure 1-b. In this research, two different types of tests, the QS compression and high velocity impact tests, were conducted for different purposes. The QS tests were performed on the specimens with 4 mm thickness to compare the stiffness of panels manufactured using Flax/PP and Flax/PPC. The impact tests were performed on the specimens with 10 mm thickness for investigating the responses of Flax/PP and Flax/PPC panels to high velocity impact loading.

2.3 Compression test

Measurements were performed using a dynamometer Instron® 5543 testing machine with 1 kN loading cell on 4 mm-thick specimens. The set-up is shown in Figure 4. The pristine composite sample was placed on a picture frame, with the same dimension as the one used in the gas-gun tests and indented with the same projectile. The picture frame was positioned using four locating pins. A 0.5 mm/s loading speed was applied.

2.4 High-velocity impact tests

The impact test specimens, with a thickness of 10 mm, were drilled around edges and then clamped around the edges using twelve ‘M8’ bolts (see Figure 1-b). The opening of metal clamp is 70 mm x 70 mm. The DIC technique was used to measure the distortion of the target. The technique is based on a stochastic pattern which was applied on the surface of the specimen [21]. The two high-speed cameras (Phantom Miro M/R/LC310) created a pair of image sequences. The algorithm used the image sequences to calculate deformation of the target. The maximum contrast was important for the test set-up. The contrast was created by painting in white with an acrylic paint and then hand-painting of the back surface of the specimen using black markers. 3-5 pixels are recommended for the process of the images sequences. In this experiment, the speckles were painted between 0.7 mm to 1.0 mm by hand painting.

A gas gun apparatus was used to achieve high velocity impacts. Helium was pressurized to 10 bars using a 4-litre cylinder. The high density polyethylene (HDPE) projectile, having a flat-fronted nose, diameter equal to 24.85 ± 0.05 mm and weight 20 ± 0.1 g, was accelerated through a 3-meter-long barrel and controlled via a pneumatic-based valve. The projectile velocity was measured by two pairs of IR sensors located at the barrel end, which connected with an oscilloscope. The set-up is shown in Figure 5 [22]. In order to view the deformation of the composite, two synchronized high-speed cameras were positioned to the rear of the target.

3. Results

3.1 Quasi-static testing results

All the investigated composite specimens are compared in Figures 6a, 6b and 6c, in terms of linear load response and stiffness, respectively.

Specifically, the PPC specimens have an average bending stiffness of $593.2 \pm 0.5\ N/mm$, which is 17.3% larger than the average value delivered by PP ones ($505.7 \pm 1.6\ N/mm$). It can be seen in Figure 6c that test results are consistent. The improved compatibility at the matrix-fibre interface induced a considerable increase of the bending stiffness of the composite samples.

3.2 High-velocity impact testing results

All impact testing was carried out using a compliant impactor (HDPE) projectile with three different impact velocity ranges (40.5 m.s^{-1}, 57.4 m.s^{-1} and 60.0 m.s^{-1}). The out-of-plane displacement maps were created using 3D DIC. The 3D DIC out-of-plane displacement maps reveal the progression of the deformation during the impact event. Note that no visible permanent deformation occurred in the samples after the impact event.

3.2.1 Flax/PP composite specimens

Figure 7 reveals the 3D DIC out-of-plane displacement results for the flax/PP composite specimens subjected to an impact velocity of 41.6 m.s^{-1}. The duration of impact was around 0.15 ms and the maximum out-of-plane displacement was observed at around 1.3 mm. The maximum out-of-plane displacement was monitored as a circle area on the out-of-plane displacement maps as seen in Figure 7.

Figure 8, shows the 3D DIC put-of-plane displacement results obtained from flax/PP composite specimens tested with a velocity of impact of 57.0 m.s^{-1}. The maximum out-of plane displacement was determined at around 2 mm. The different level of out-of-plane displacement was observed clearly as a ring on the out-of-plane displacement maps.

Figure 9 depicts the out-of-plane displacement maps attained from flax/PP composite specimens tested with 60.0 m.s^{-1} impact velocity, under loading and unloading phase and, again, a maximum out-of-plane displacement of 2 mm was noted.

3.2.2 Flax/PPC composite specimens

Figure 10 shows the out-of-plane displacement contours under impact tests at 39.3 m·s^{-1} during loading/unloading steps with an impact velocity being 39.3 m.s^{-1}. Figure 11 depicts results of impact measurements performed on flax/PPC composite specimens under energy of impact of 33.41 J. The maximum out-of-plane displacement was determined at around 1.9 mm. The difference from the out-of-plane displacement was clearly seen as a circle.

Finally, Figure 12 refers to impact of flax/PPC composite specimens at 60 m.s^{-1}. Results showed a maximum out-of-plane displacement equal to approximately 2 mm. In this case, the duration of the impact event is around 0.175 ms and all kinetic energy was absorbed after 0.2 ms.

3.3. SEM Analysis

There was no visible damage on all the specimens so it was not possible to compare damage within these specimens. The results from DIC was compared to understand the effect of the coupling agents. Specimens were intentionally fractured and SEM observations were performed. Specifically, the examined surfaces were obtained by fracture of a specimen, cut from a laminate of each material in liquid nitrogen to avoid yield bands from ductility of the polypropylene matrix, which can hide characteristic morphological details of the interface.

Figure 13 shows the comparison of the fractured surfaces without (A) and with (B) coupling agent. In the absence of the compatibilizer (Figure 13-A), the lack of enough interfacial affinity between the reinforcement and matrix causes matrix fracture, hence showing the structure of the intact flax fibre fabric. In the compatibilized system, represented by the SEM micrograph of the picture on the right, the reinforcement is apparently adequately anchored to the surrounding matrix such that both the fibre and matrix fracture together. This indicates a better quality of the composite with respect to fracture due to an improved bond between the reinforcement and matrix. The corollary is that better interfacial adhesion arising due to the coupling agent resulted in an improved composite with respect to impact properties such that if fracture initiates, it occurs simultaneously in both the fibre and matrix that is highly desired. This alludes to the enhanced impact performance of the Flax/PPC composite.

3.4 Discussion

In this study, effects of a polypropylene grafted maleic anhydride coupling agent, commonly used for polypropylene based composite systems, on the QS compression and high velocity impact behaviour of flax fibre reinforced composites have been studied.

In mechanical tests undertaken under static conditions, it was found that the bending stiffness of flax/PPC composite specimen is higher than the flax/PP composite specimen. This effect can be explained considering that the action of the punch consists of subjecting the specimen locally to a bending load with a stiffness subjected to two modes of loading, i.e. upper surface compression and bottom surface tension.

In this case, a key role is played by the interfacial bonds at the fibre/matrix interface: the greater the adhesion between consecutive layers of plastic and reinforcement, the smaller the reciprocal sliding [23]. Thus, the improved bending stiffness found in this study is a sign of efficient action of the additive included in flax / PPC specimens.

With regards to the impact behaviour, the out-of–plane displacement of the composite specimens increases with the impact energy but, under the same energy parameter, it is reduced by 11-22% in presence of the coupling agent (Figure 14). Figure 14 also confirms, via the observed linearity, that the impact energy is proportional to the out-of-plane displacement. This is a corollary of the fundamental expression that work done is the force by the distance moved by the force.

In fact, as the interfacial adhesion increases, the composites respond to applied loads with a behavior that increasingly reflects the performance of the more rigid reinforcing phase. For this reason, it is possible to state that the coupling agent reduces the plastic deformations (out of plane displacements) and, consequently, it decreases the elastic energy absorbed during the loading, under the same impact energy.

A lower out-of-plane displacement incorporating a coupling agent for a given impact loading bodes well for protection of internal circuitry and mechanical components within drones and automobiles. It is anticipated that this advantage will hold at even higher impact energies.

4. Conclusions

The effect of the addition of a coupling agent in flax/polypropylene composites was studied under both compression and high velocity impact loading. The comparison of impact performances was implemented using the 3D Digital Image Correlation (DIC) maps of out-of-plane displacement.

The coupling agent improves the stiffness and induces a reduction of the out-of-plane displacement of flax composites subjected to impact loading: both these effects demonstrate an efficient behaviour of the included additive with a significant improvement in the interfacial adhesion.

In conclusion, lightweight flax/PP composites including a beneficial coupling agent may offer interesting perspective of applications thanks to their improved mechanical properties and higher sustainability with respect to conventional reinforced systems. Potential applications include aviation drones and automobiles. Further testing at other impact velocities can serve to validate the results from this study.

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Fig. 1 Composite investigated: (a) 2x2 twill flax fabric and (b) image of clamped flax composite by twelve “M8” bolts

Fig. 2 Processing conditions used to prepare flax/PP fabric laminates

Fig. 3 Drawing of each specimen (all dimension in mm)

Fig. 4 QS compression test: (a) Schematic and (b) photograph of experimental set-up

Fig. 5 Schematic of gas gun with Three Dimensional (3D) DIC

Fig. 6 Quasi-static experiments: (a) Load-displacement curves and (b) & (c) average bending stiffness measured from samples with and without coupling

Fig. 7 3D DIC out-of-plane displacement results for flax/PP composite specimens without coupling agent tested during the impact event (impact velocity and energy: 41.6 m.s^{-1}, 17.3 J)

Fig. 8 3D DIC out-of-plane displacement results for flax/PP composite specimens without coupling agent tested during the impact event (impact velocity and energy: 57.0 m.s^{-1}, 32.3 J)

Fig. 9 3D DIC out-of-plane displacement results for flax/PP composite specimens without coupling agent tested during the impact event (impact velocity and energy: 60.0 m.s^{-1}, 35.2 J)

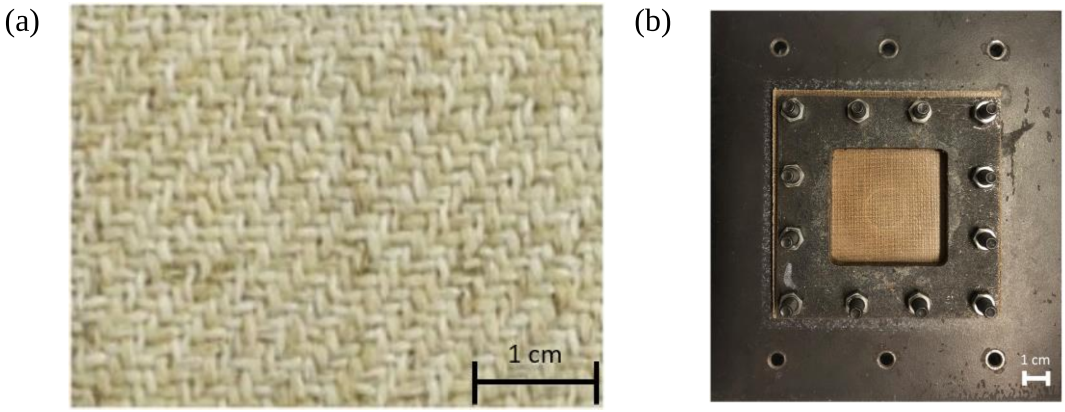
Fig. 10 3D DIC out-of-plane displacement results for flax/PPC composite specimens with coupling agent tested during the impact event (impact velocity and energy: 39.3 m.s^{-1}, 15.4 J)

Fig. 11 3D DIC out-of-plane displacement results for flax/PPC composite specimens with coupling agent tested during the impact event (impact velocity and energy: 57.8 m.s^{-1}, 33.4 J)

Fig. 12 3D DIC out-of-plane displacement results for flax/PPC composite specimens with coupling agent tested during the impact event (impact velocity and energy: 60.0 m.s^{-1}, 36.2 J)

Fig. 13 SEM pictures without coupling agent (A) and with coupling agent (B) of fractured specimens. 200X magnification at 30kV.

Fig. 14 Comparison of impact energy versus out-of-plane displacement of flax/PP without coupling agent and flax/PPC composite with coupling agent



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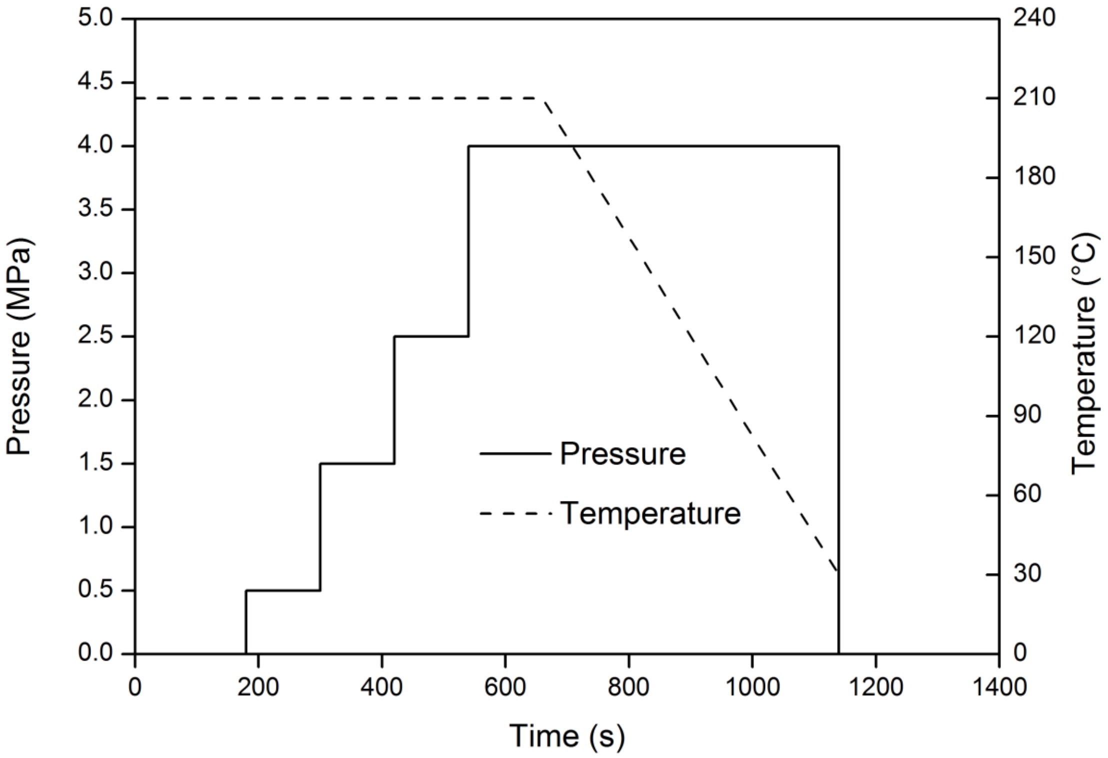

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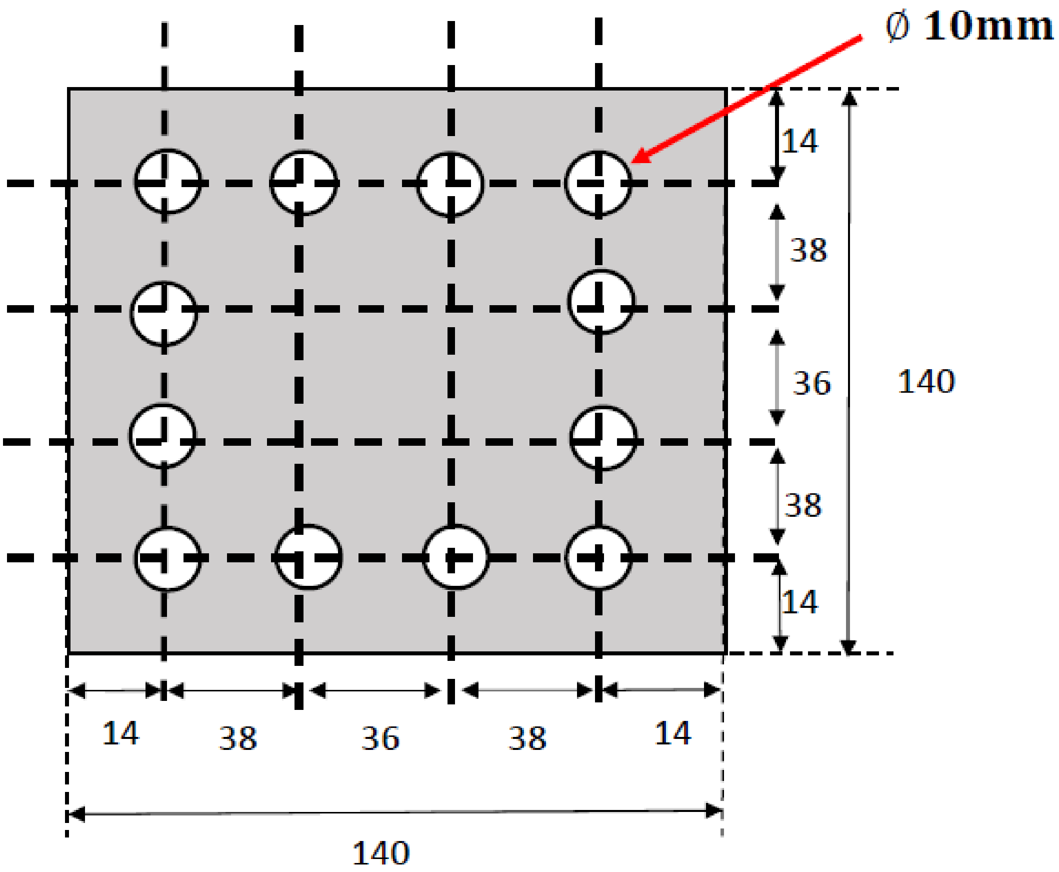
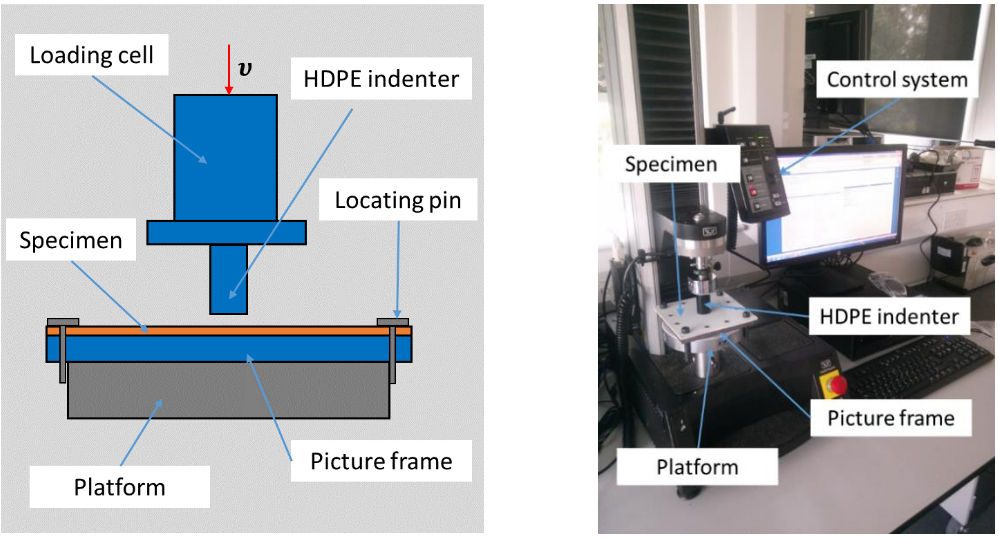


Fig. 3 Drawing of each specimen (all dimension in mm)



(a) (b)

Fig. 4 QS compression test: (a) Schematic and (b) photograph of experimental set-up

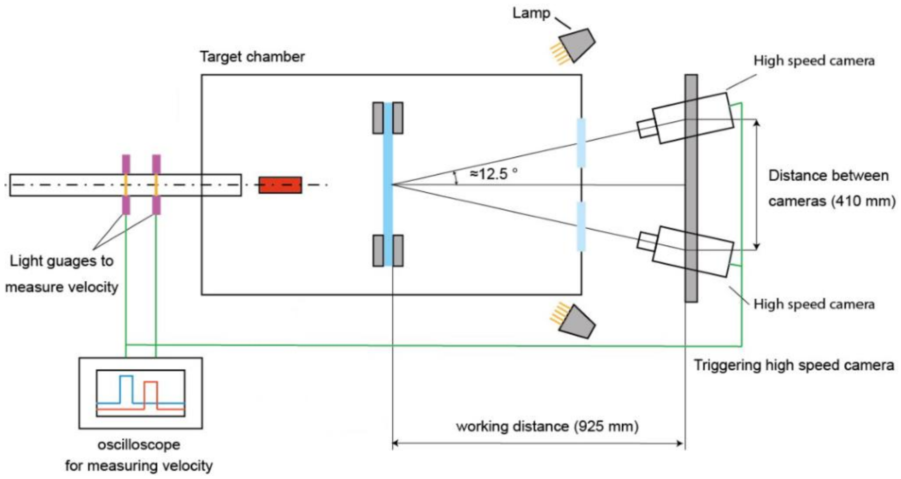
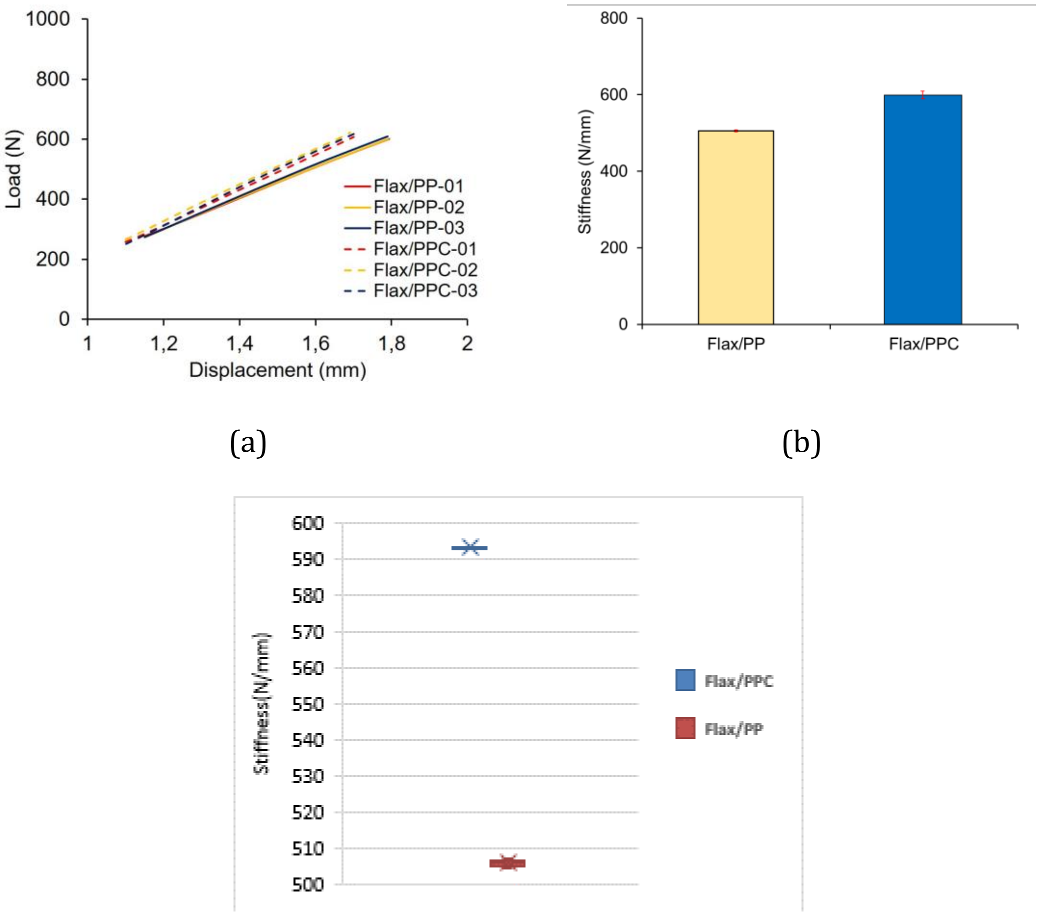


Fig. 5 Schematic of gas gun with Three Dimensional (3D) DIC



(a) (b)

(c)

Fig. 6 Quasi-static experiments: (a) Load-displacement curves and (b) & (c) average bending stiffness measured from samples with and without coupling agent

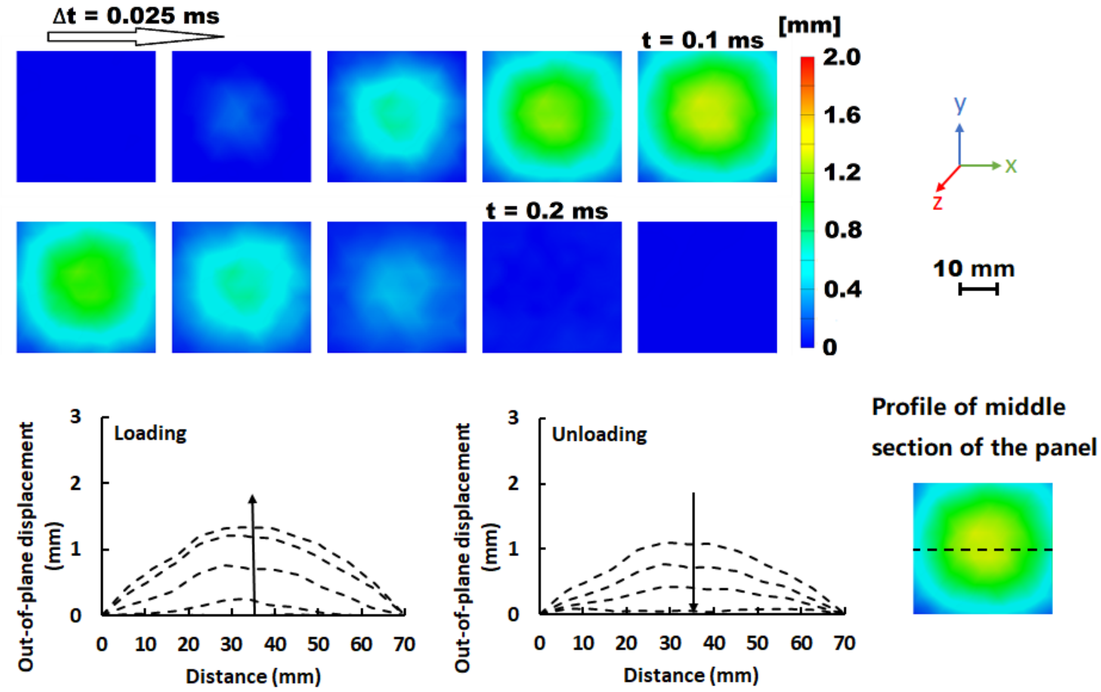


Fig. 7 3D DIC out-of-plane displacement results for flax/PP composite specimens without coupling agent tested during the impact event (impact velocity and energy: 41.6 m.s^{-1}, 17.3 J)

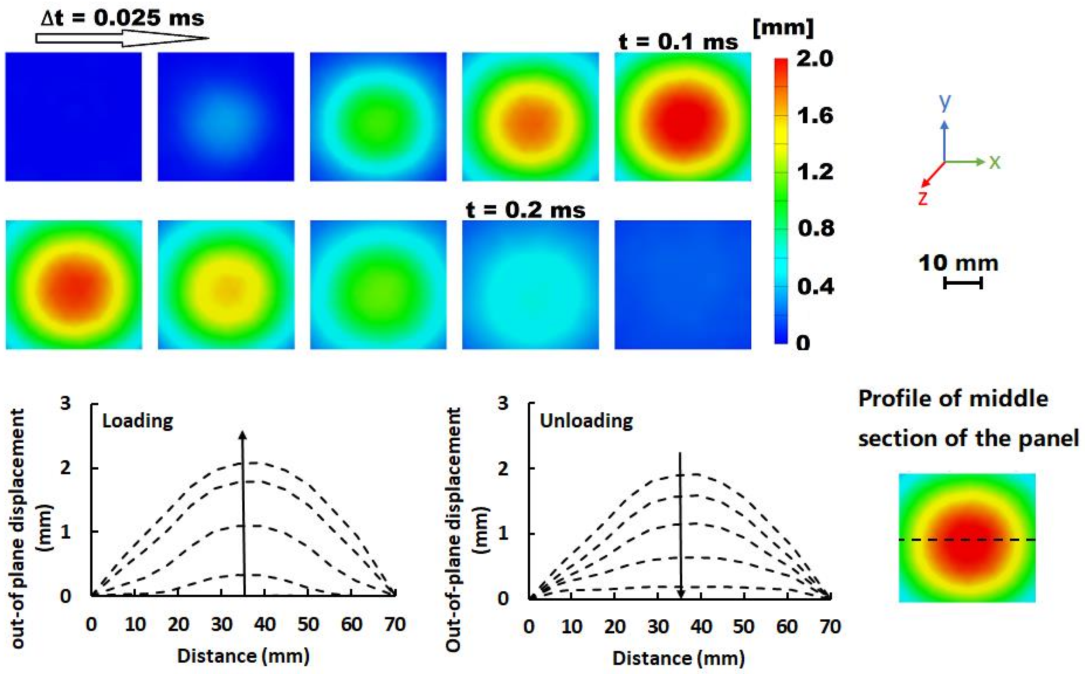


Fig. 8 3D DIC out-of-plane displacement results for flax/PP composite specimens without coupling agent tested during the impact event (impact velocity and energy: 57.0 m.s^{-1}, 32.3 J)

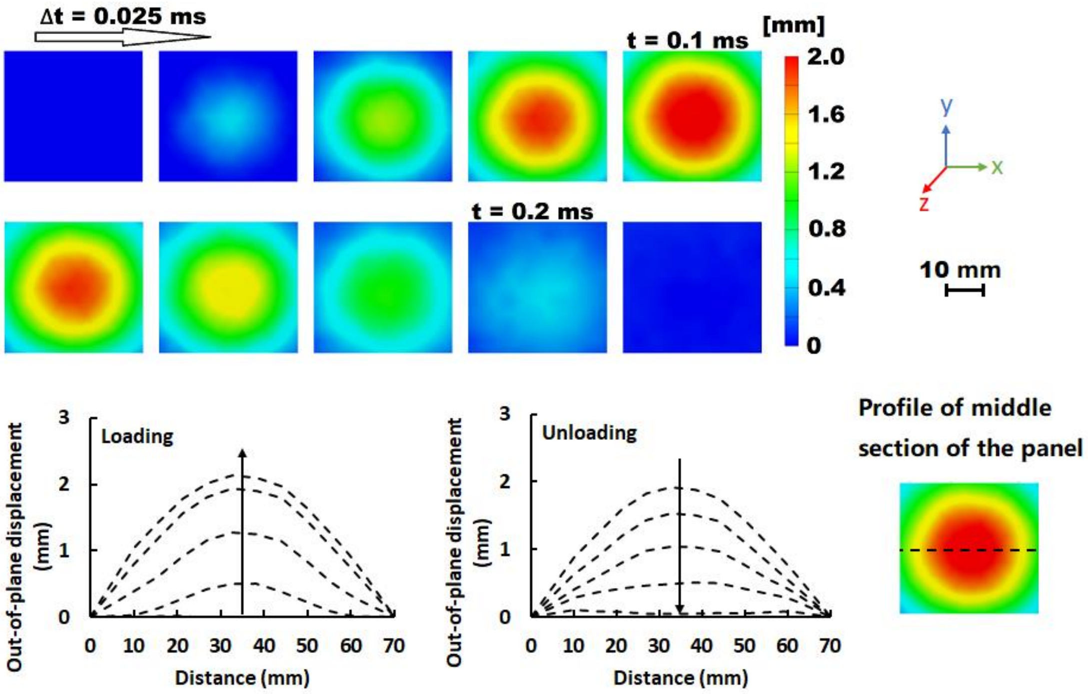


Fig. 9 3D DIC out-of-plane displacement results for flax/PP composite specimens without coupling agent tested during the impact event (impact velocity and energy: 60.0 m.s^{-1}, 35.2 J)

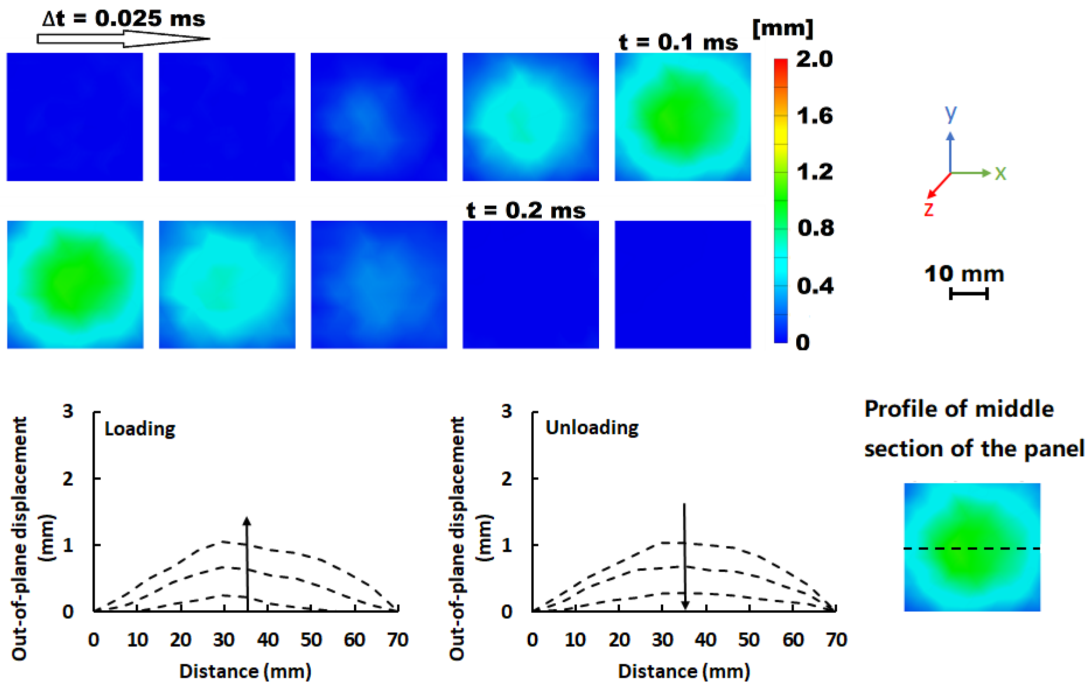


Fig. 10 3D DIC out-of-plane displacement results for flax/PPC composite specimens with coupling agent tested during the impact event (impact velocity and energy: 39.3 m.s^{-1}, 15.4 J)

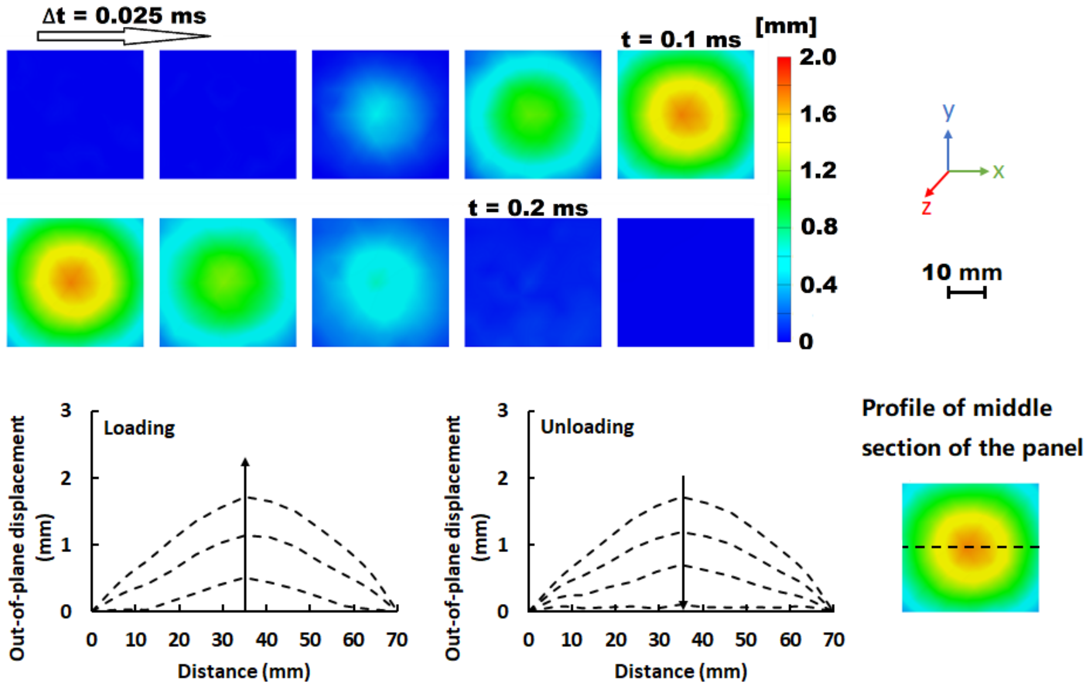


Fig. 11 3D DIC out-of-plane displacement results for flax/PPC composite specimens with coupling agent tested during the impact event (impact velocity and energy: 57.8 m.s^{-1}, 33.4 J)

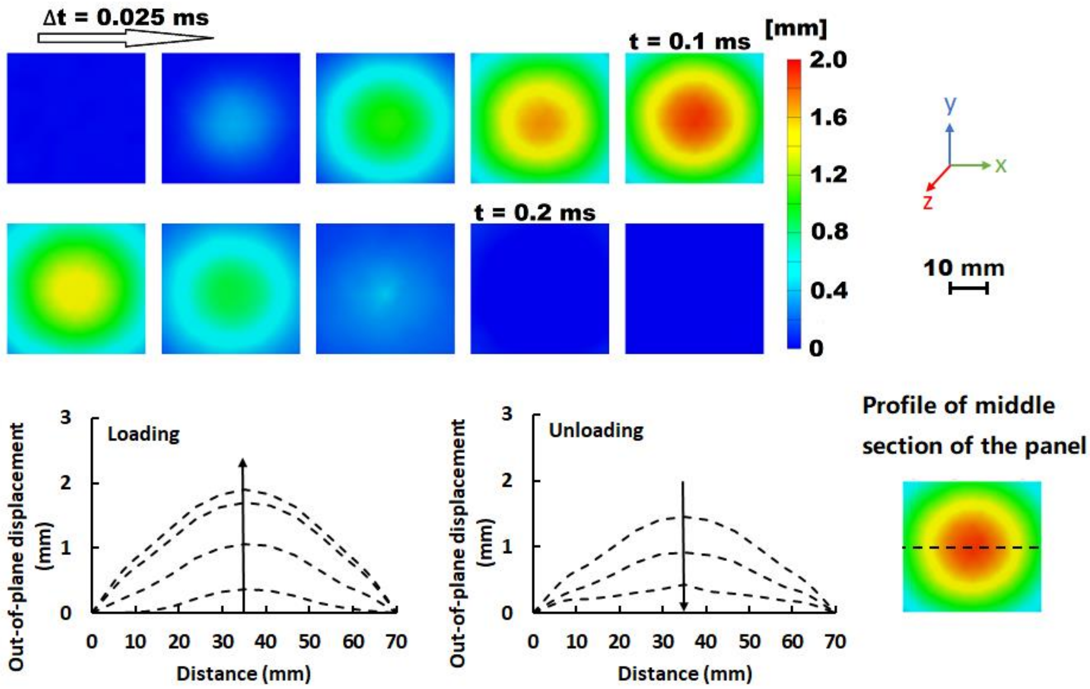
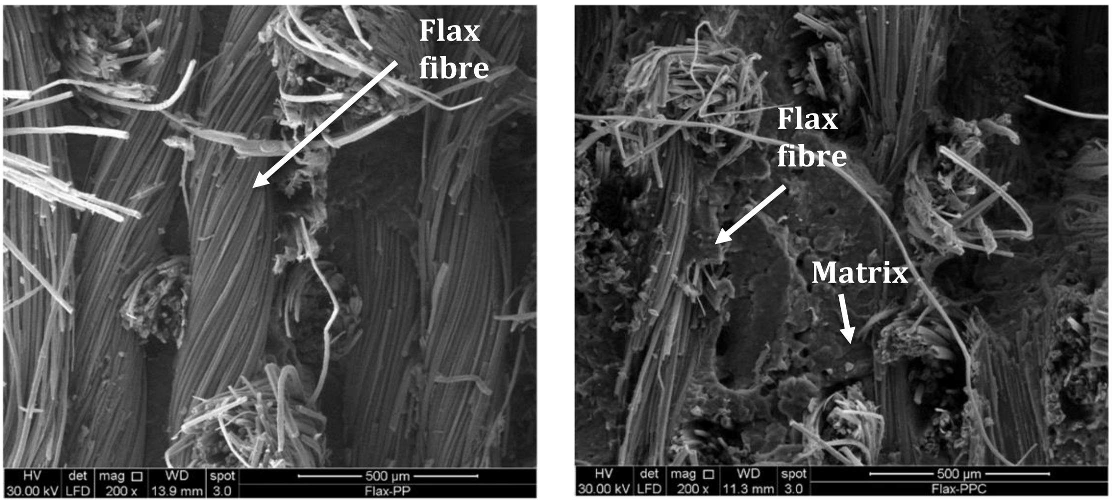


Fig.12 3D DIC out-of-plane displacement results for flax/PPC composite specimens with coupling agent tested during the impact event (impact velocity and energy: 60.0 m.s^{-1}, 36.2 J)



(A)

(B)

Fig. 13 SEM pictures without coupling agent (A) and with coupling agent (B) of fractured specimens. 200X magnification at 30kV.

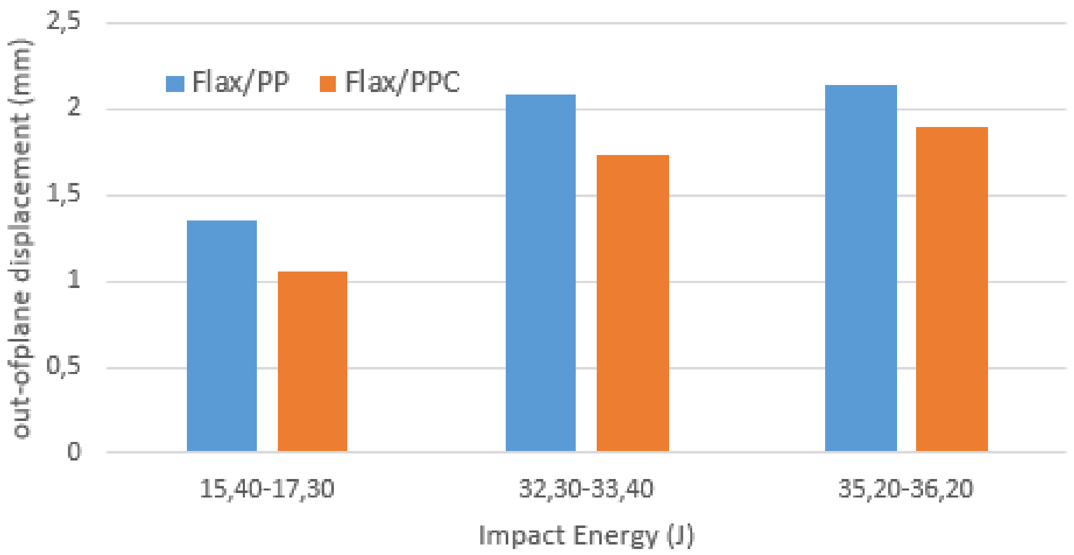


Fig. 14 Comparison of impact energy versus out-of-plane displacement of flax/PP without coupling agent and flax/PPC composite with coupling agent