COMPARATIVE STUDY ON THE IMPACT BEHAVIOR AND DAMAGE TOLERANCE OF WOVEN CARBON FIBER REINFORCED THERMOPLASTIC– AND THERMOSETTING– COMPOSITES

B. Vieille[1*], V. M. Casado[2], C. Bouvet[2]

[1]*INSA Rouen – Groupe de Physique des Matériaux, UMR 6634 CNRS, 76801 St Etienne du Rouvray*
[2]*Université de Toulouse, INSA, UPS, Mines Albi – ICA/ISAE, 10 av. E. Belin, 31055 Toulouse cedex 4*
**benoit.vieille@insa-rouen.fr*

Keywords: impact behavior, damage tolerance, thermosetting, thermoplastic

Abstract

The purpose of this study is to compare the response of TS-based (epoxy) and TP-based (PPS or PEEK) laminates under low velocity impacts. In order to assess the development of internal damage caused by the different impact energies, non-destructive examinations were done. Impact tests created pyramidal-shaped damage at the center of the specimen, extensive delamination and fiber breakages inside laminates, as well as inter laminar and intra-ply damage. CAI tests showed that PEEK-based composites display the best performance, and the compressive residual strengths of C/PPS and C/Epoxy are the lowest for every impact energy. It turns out that the three materials display a rather similar behavior at increasing impact energy as delaminated areas are larger in C/Epoxy and C/PEEK, whereas the PPS matrix toughness contributes to a less extended delaminated area. One reason for the disappointing residual strength of TP-based composites may be the constraint on the development of the plastic deformation zone in the resin rich area between the woven-plies and at the fiber bundles crimp, preventing the matrix toughness to give to the laminates a better damage tolerance.

1 Introduction

Polymer matrix composite laminates are prone to delamination when impacted. This behavior generally results in a low damage tolerance, which is of great concern for load carrying applications. To discuss the impact behavior of polymer matrix composites it is helpful initially to consider the nature of constitutive materials, as well as the nature of the reinforcement [1]. In composite materials, as well as in unreinforced materials, the use of thermoplastics (TP) predominates, although their use is usually restricted to short fiber reinforced injection moldable formulations. Long and continuous fiber reinforced composites are still dominated by thermosetting (TS) polymer matrices. In long and continuous fiber reinforced composites, TS are particularly suited for impregnation into the reinforcing fibers by manual or semi-automated means. Thus, TS matrix composites have been widely used for aeronautical applications over the last four decades. However, TS-based composites show some issues in their manufacturing process (low-temperature storage, long curing cycles, irreversible process…). Nowadays, high-performance TP matrix composites (e.g. PEEK and PPS) present promising alternative to those drawbacks [2]. Crystallinity in high-performance polymers is also important, because it has a strong influence on both chemical and mechanical properties: the crystalline phase tends to increase stiffness and tensile strength, while the amorphous phase is more effective in absorbing impact energy. Thus, there is a strong current trend towards a greater use of TPs in high performance composites structures, driven by considerations of mass reduction as well as tackling issues of sustainability and recyclability [3]. TP-based composites are already well established within aircraft interiors but airframe components in primary structure from this class of material are just emerging. This progression of TP resins from secondary to primary structure is opening up the design and manufacturing envelope with a new set of production characteristics. There are numerous engineering reasons why TP composites are attractive as aerostructures, such as increased toughness compared with TS alternatives and inherent flame retardancy. At the same time, low-cost processing is obtained by using manufacturing processes like thermofolding, stamping, welding and co-consolidation.

2 Literature review

Since low velocity impact being one of the most detrimental solicitations for laminates, high-performance TP are considered in composites structures mainly for damage tolerance reasons. Impact-induced damage is particularly critical because it drastically reduces the residual mechanical properties of the structure [4-6]. A few authors have compared the impact behavior of TS- and TP-based composite structures, and their effects on residual strength [7-12], as well as the damage tolerance of UD-ply and woven-ply laminates [12-14]. The following literature review is not aimed at giving a general overview of the impact behavior of TS-based laminates for which a great number of references are proposed in the literature [4]. However, it focuses on the comparison of the specific impact performance and damage tolerance of polymer-based laminates depending on the matrix nature (TS or TP). Early in the 90s, the impact performance and damage tolerance of TP-based composites have been studied in order to understand why such materials are often more damage tolerant than TS-based composite materials. In this aim, a few authors have investigated the influence of examined matrix type and morphology on the ability of TP-based composites to withstand penetration, absorb energy and sustain damage at different temperature levels. Most of the studies about the impact performance and damage tolerance of TP-based composites deals with PEEK-based composites [10-11][15-18]. However, only very few references report the impact behavior of PPS-based laminates [9][15][19-21], and it appears from the literature results that they display a better resistance to impact damage than Epoxy-based composites. The effect of the impact velocity is found to be insignificant, whereas the impact energy significantly affected the impact performance of the laminates. C/PPS UD-ply laminates evinced a high resistance to perforation byway of extensive delamination. C/PEEK UD-ply laminates showed an ability to confine the damage zone and hence, to markedly increase the damage tolerance of the laminates. Thus, higher Compression After Impact (CAI) strengths are generally observed in C/PEEK compared to C/Epoxy, and the reasons have already been explained [15]. The process of delamination propagation in the final stage of CAI tests performed on C/Epoxy UD-ply laminates is well understood: transverse delamination penetration to the loading direction causes buckling deflection reverse in the impact side and reduces the load carrying capacity of the delaminated laminates. In addition, comparatively to UD-ply composites, woven-fabric composites are characterized by better impact resistance, damage tolerance and high toughness [20]. The purpose of this work is to examine the specific contribution of the matrix to both impact performance and damage tolerance of different types of PMCs. In this aim, low velocity impact tests have been carried out for different impact energies, and fractography/C-scan analysis have been performed so that the damage mechanisms and the delaminated area during impact can be identified. At last, in order to assess the severity of damage, CAI tests have been performed.

3 Materials and experimental procedure

The composite materials studied in this work are carbon fabric reinforced prepreg laminated plates consisting of different matrix: TP (PPS or PEEK) and TS (Epoxy). The PPS resin (Fortron 0214) is supplied by Ticona, the PEEK resin (grade 150) is supplied by Victrex, and the epoxy resin (914) is supplied by Hexcel. The woven-ply prepreg laminate consists of 5-harness satin weave carbon fiber fabrics whose reference is T300 3K 5HS, and is supplied by SOFICAR. The volume fraction of fibers is 50%. The prepreg plates are hot pressed according to a Quasi-Isotropic lay-up: [(0,90)/(±45)/(0,90)/(±45)/(0,90)/(±45)/(0,90)]. The specimens were cut into flat panels with a water-cooled diamond saw. Specimens are 100x150mm² plates, and complies with the standard Airbus AITM 1-0010, except for the recommended thickness (4 mm). The laminates' thickness was averaged from measurements at different points: 2.25 mm for C/PEEK, 2.29 mm in C/PPS, and 2.4 mm in C/Epoxy laminates. Low velocity impact tests have been carried out at room temperature on the three composite materials using a drop tower (steel hemispherical indentor diameter = 20 mm – impactor weight = 2.077 kg), for different impact energies ranging from 2 J ($v_{indentor} \approx 1.4$ m/s) to 25 J ($v_{indentor} \approx 5$ m/s) such as: 2J, 6J, 10.5J, 17J, 25J. Two specimens have been tested in each configuration. According to the previous standard, the BVID (Barely Visible Impact Damage) is defined by 0.6mm of indentation after relaxation of the structure and without being exposed to any humidity. The data acquisition is achieved using a Yokogawa DL708 digital oscilloscope, which measures three types of signals: force, intensity of the laser beam and displacement. The frequency range for data acquisition is set to 200 kHz for the three signals. A C-Scan non-destructive control has been carried out in order to examine the damage pattern. Even though they are not presented here, microscopic observations have been performed to get information about failure mechanisms. At last, CAI tests were performed using a Schenk hydraulic testing machine at room moisture, and displacement-controlled rate (0.2 mm/min). Because of the low thickness of the studied laminates, the standard's boundary conditions of the CAI tests have been modified (70*50mm) in order to increase the calculated buckling strengths (See Fig. 4b).

4 Results

For every impact energy, it appears that C/Epoxy presents the highest ratio of dissipated energy compared to impact energy, whereas the energy dissipated during impact is virtually the same in C/PPS and C/PEEK (see Tab. 1). Such a ratio usually increases to reach a maximum value at the onset of perforation [21], as it can be observed in the C/Epoxy case. The first energy causing an indentation below the BVID is 10.5J for an Epoxy specimen.

	C/PEEK					**C/PPS**					**C/EPOXY**				
E_{impact} (J)	1.88	5.92	10.25	16.95	23.65	1.73	5.78	10.37	16.9	24.4	1.75	5.85	10.3	17.4	25
Max. displacement (mm)	2.61	4.66	6.19	8.59	11.36	2.5	4.44	6.31	8.34	11.35	2.42	4.55	6.47	9.9	Perforation
$E_{dissipated}$ (J)	0.45	3.44	6.74	13.48	20.87	0.3	2.96	6.79	13.05	21.94	0.64	3.7	7.78	16	22.6
$E_{dissipated}/E_{impact}$ (%)	24	58	66	80	88	17	51	65	77	90	37	63	76	92	90

Table 1. Results of impact tests for different impact energies

The force-time curves show the duration of the contact between indentor and specimen surface, the maximum force reached and the appearance of damage (See Fig. 1). The force-displacement curves give the specimen's stiffness (slope of the curve), the maximum displacement and some information about damage onset (see Fig. 2). The first type of damage is matrix cracking, which does not significantly change the overall stiffness of laminates. However, matrix cracks tips may act as onset sites for delamination and fiber breakage which do change the local stiffness of laminates [1]. In addition, the specimen can absorb the impact energy by other means including indentation (representative of local matrix crushing and local fiber breakage), delamination, splitting or fibers peeling on the non-impacted side. For TP-based laminates, the onset of the first damage can be observed at around 0.5ms. There is an almost sinusoidal increase until the first significant damage appears, causing a sudden drop of the slope (approximately 1ms in TP specimens and 0.5ms in C/Epoxy specimens). It is followed by an oscillating increase until maximum force, at approximately 3ms for the 2J impact, and earlier for others impact energies. The higher the impact energy is, the earlier the maximum force is reached – the 17J impact reaches the maximum force at 2ms, at virtually the same time than the 25J impact. After this peak, the force decreases almost steadily until zero. A threshold seems to appear at about 3kN for 10.5J, 17J and 25J impact energies. The 2J impact does not cause any damage to specimens, whereas the 25J impact induces the highest damage. At 6J, some fiber breakages and the initiation of a longitudinal crack appeared. Most of the impact events are completed at about 7ms, but the 25J test is longer because of the significant penetration of the indentor into the laminate. Finally, the maximum force reached in C/PPS is always equal or slightly higher than the one reached in C/PEEK. C/Epoxy laminates display similar performance to TP composites, with two main differences: the force threshold is lower (2.5kN), and the 25J impact causes specimen's perforation, hence justifying a residual force at the end of the test. For a relevant comparison of the impact performance of the three materials, force-displacement curves have been compared for every impact energy (see Fig. 2). C/Epoxy laminates present the lowest impact resistance at 6J, and suffer more damage than TP-based laminates, although the maximum displacement is similar. For higher energies, maximum displacement is always higher for C/Epoxy specimens. The onset of cracks along warp/weft directions is observed on the non-impacted side at 10.5J.

(a) (b)

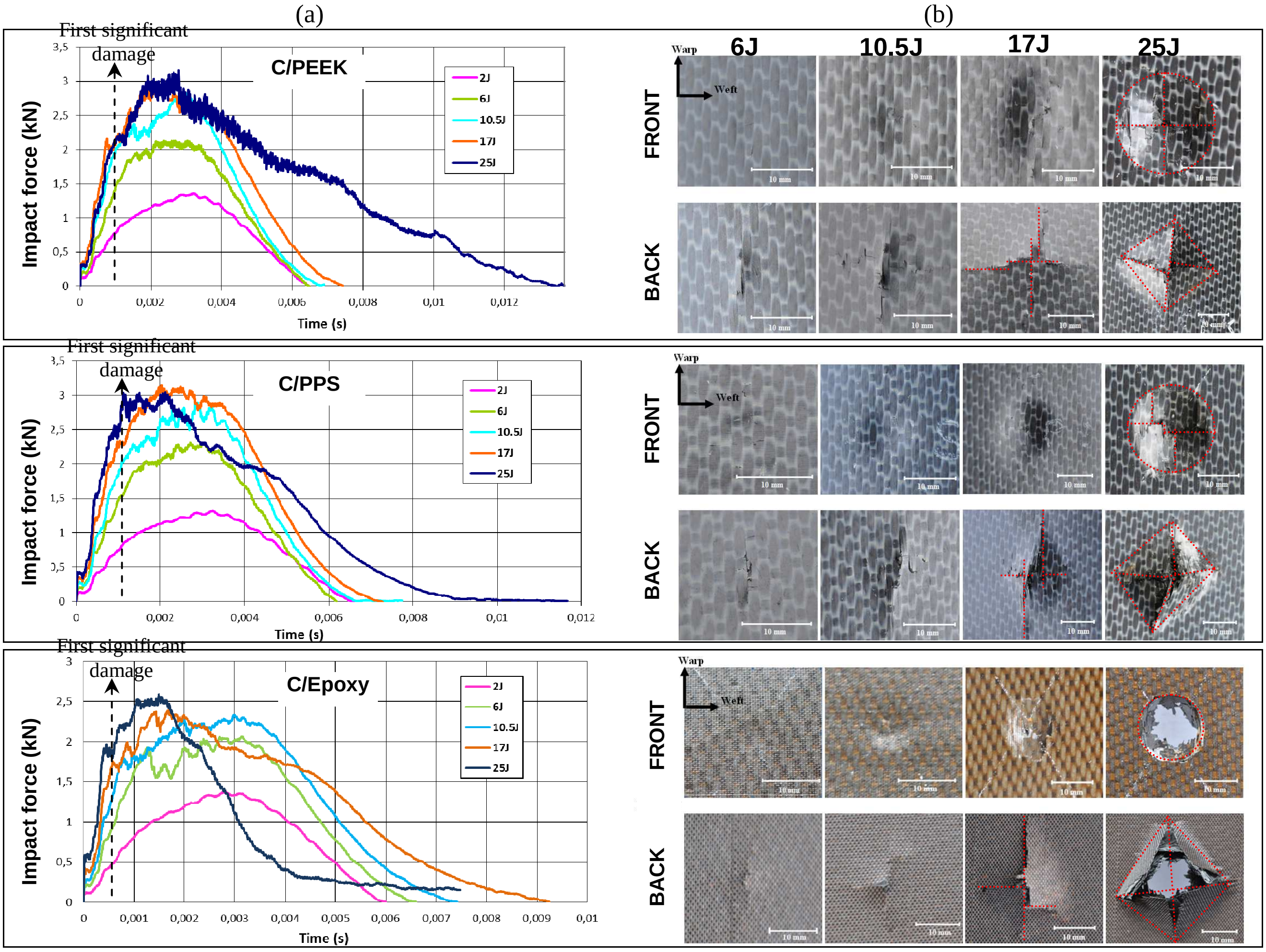

Figure 1. Load-time histories of the different impact energies carried out on each material

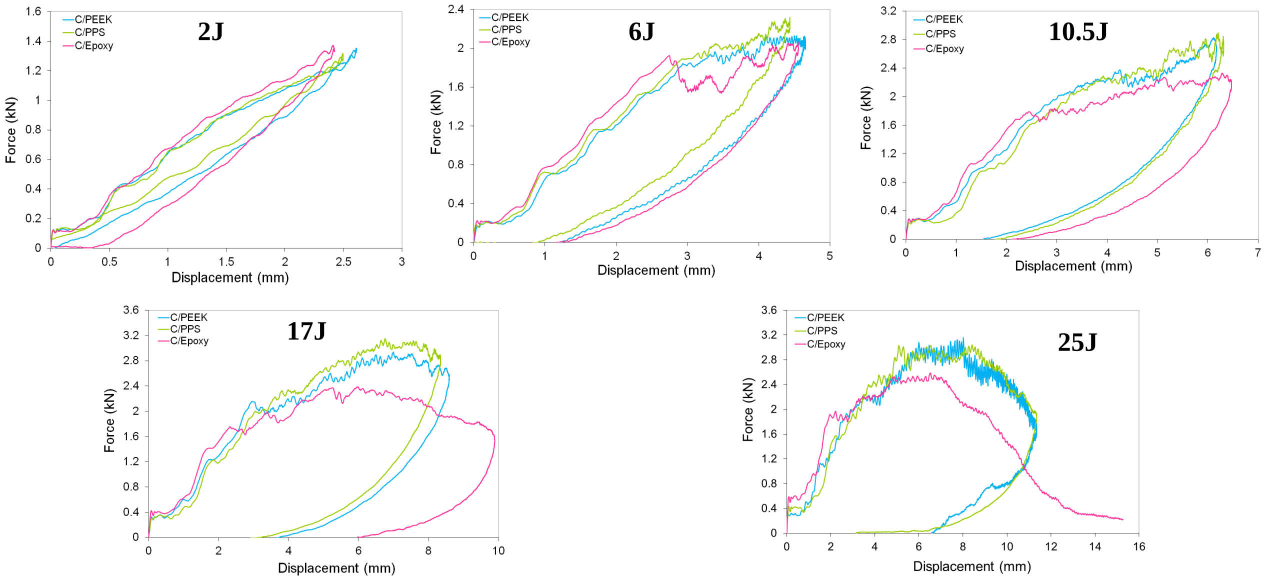

Figure 2. Comparison of load-displacement curves for every impact energy

At 17J the hemispherical indentation on the impacted side is clear (this is the first energy that creates an indentation over the BVID for TP specimens), and fiber splitting start being quite large, while cracks along warp/weft directions significantly developed on the non-impacted side. Lastly, the 25J impact seriously penetrates C/PEEK and C/PPS laminates. At 17J, penetration occurs earlier, and the 25J impact perforates C/Epoxy laminates (see Fig. 1b). TP-based laminates show a cross-shape failure on the non-impacted side, and cracks propagate along directions slightly oriented with respect to warp/weft directions (see Fig. 1b). On the non-impacted side of C/Epoxy laminates at 17J, there are two cracks along the weft direction, and one crack along the warp direction. The delaminated area, calculated from C-scan observations of impacted specimens (See Fig. 3), is related to the dissipation of energy during the impact and the residual strength of laminates. C/Epoxy laminates display larger delaminated areas than TP composites, reaching values twice higher for the same impact energy, except for the 25J impact for which the specimen was perforated (See Fig. 4a). The delaminated area is considerably more extended in C/PEEK than in C/PPS laminates at high impact energy. At last, the residual compressive strengths of C/PEEK laminates is higher for every impact energy (See Fig. 4b), whereas the compressive strength of non-impacted C/Epoxy laminates (758 MPa) is about 10% (C/PEEK) and 45% (C/PPS) higher, respectively [22]. The calculated buckling strengths (See Fig. 4b) are virtually equal in TP-based laminates (about 320 MPa), but higher in C/Epoxy laminates (about 420Mpa). For a 2J impact, the residual compressive strength is 20% and 50% higher in C/PEEK in comparison with C/Epoxy and C/PPS laminates respectively. For a 25J impact, the residual strength is virtually the same in C/PEEK and C/Epoxy, and 12% higher than C/PPS one.

5 Discussion

At high impact energies, the impact damage is represented by a delamination edge geometry whose shape is pyramidal. The damage area is similar on both impacted/non-impacted sides of TP-based laminates (see Fig.1b). However, the TS-based laminates were relatively free to delaminate on the non-impacted surface as a result of the longitudinal splitting of fibers along warp/weft directions [13]. There is also a clear indication that, on the impacted surface, the damage area was generally larger for TS laminates than for TP laminates, probably because of the extensive splitting on the non-impacted surface of TS laminates (see Fig. 3). In TP composites, the higher impact-damage resistance of TP laminates is apparently related to their high inter-laminar and intra-laminar fracture resistance [20]. It seems relevant here to recall the GIc fracture toughness for neat TP resins (0.7 kJ/m² for PPS – 4 kJ/m² for PEEK), and the delamination GIc for 24 plies fabric laminates (0.9 kJ/m² for C/PPS – 2.1 kJ/m² for C/PEEK). The ductile matrix of TP laminates is associated with an inherently tough weave structure.

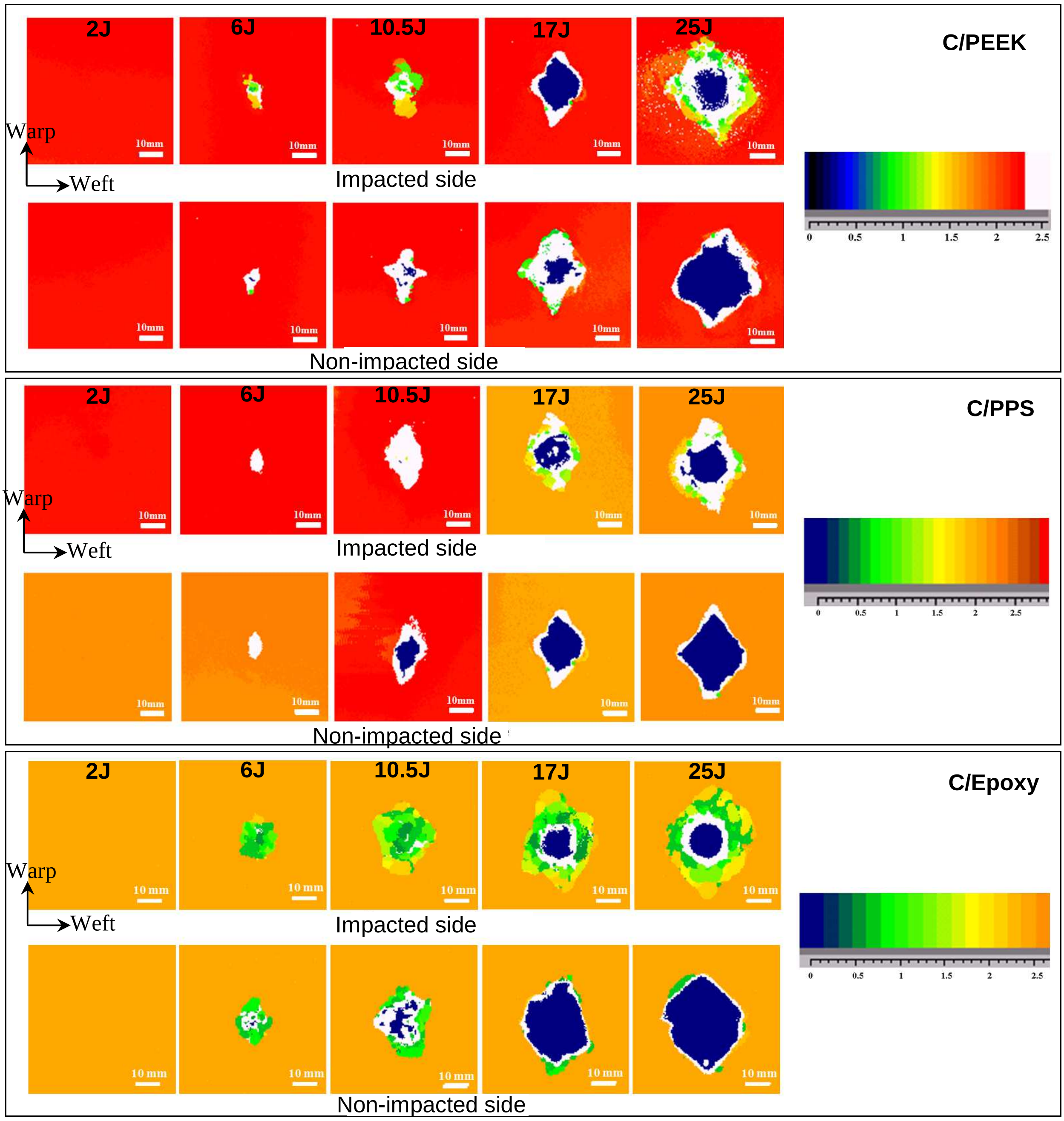

Figure 3. C-Scan control of impacted specimens: changes in damage pattern as a function of impact energy

Both features seem to play a significant role in the residual strength after impact, because they are particularly beneficial in limiting the propagation of longitudinal cracks. On 25J impacted laminates and with respect to buckling strengths (See Fig. 4b), the compressive strength decreases by 51% (C/PEEK), 56% (C/PPS) and by 62% in TS-based laminates. These results suggest that PPS matrix toughness [22] contributes to the inhibition of fiber splitting in weave structure leading to large stress concentrations near the resin-rich regions, hence justifying less extended delaminated areas in C/PPS laminates (see Fig. 4a). However, even though the weave structure and fiber bundles crimp effectively limit an excessive delamination at increasing impact energy, they also limit the development of plastic deformation zones in the resin rich area between the woven-plies and at the fiber bundles crimp. Thus, the delaminated area is larger in C/Epoxy and C/PEEK than in PPS-based laminates. Such a delamination is ultimately detrimental to buckling instabilities under CAI loadings. The previous results are in agreement with the CAI strength of quasi-isotropic woven-ply C/PEEK laminates, which remained at 47% of the value for virgin material after sustaining a 29J impact [15].

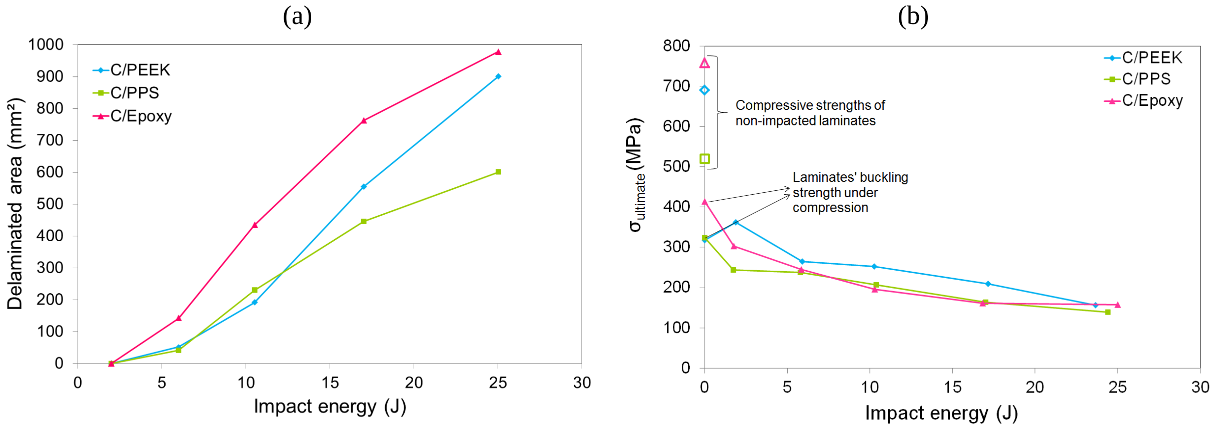

Figure 4. Comparison of impact behavior and damage tolerance depending on the impact energy: (a) Delaminated areas resulting from C-Scan results – (b) Residual CAI strengths

6 Conclusion

Among the studied materials, C/Epoxy laminates presents the worst response under low velocity impact, showing larger delaminated areas. TP-based laminates subjected to impacts are characterized by a reduced damage (particularly for C/PPS). These results confirm that a tougher matrix can be associated with better impact performance of the corresponding composite system. For TP-based laminates, the BVID is reached at 17 J of energy, whereas it is reached at 10.5 J for C/Epoxy. In addition to the nature of the matrix, the reinforcement weave structure limits extensive growth of delamination, but fiber breakages are more common and appear at lower impact energies because of fiber crimps. The features and advantageous failure mechanisms are identified: inherent toughness of the fabric; the availability of resin-rich regions at the fiber bundles crimp where plastic deformation can develop (in C/TP); crack propagation along the undulating pattern of the yarns creating a large fracture surface area; and multiple crack delamination on the impacted side. From CAI tests results, PEEK-based composites display the best performance, and the compressive residual strengths of C/PPS and C/Epoxy are the lowest for every impact energy. It also turns out that the three materials display a rather similar behavior at increasing impact energy as delaminated areas are larger in C/Epoxy and C/PEEK, whereas the PPS matrix toughness contributes to a less extended delaminated area. One reason for the disappointing performance of TP-based composites may be the constraint on the development of the plastic deformation zone in the resin-rich regions, preventing the matrix toughness to give laminates a better damage tolerance.

References

[1] Hancox N.L. *An overview of the impact behavior of fibre-reinforced composites* in "Impact behavior of fiber-reinforced composite materials and structures", edited by Reid S.R. and Zhou G., CRC Press, Woodhead Pub. Ltd (2000).

[2] Offringa A.R. Thermoplastic composites – rapid processing application. *Composite Part A*, **27**, pp. 239-236 (1996).

[3] Spruiell J.E., Janke C.J. *A review of the measurement and development of crystallinity and its relation to properties in neat PPS and its fiber reinforced composites* in “Tech. report, Metals and Ceramics Division”, US Dept. of Energy (2004).

[4] Cantwell W.J., Morton J. The impact resistance of composite materials - a review. *Composites*, **22(5)**, pp. 347-362 (1991).

[5] Jang B.P., Kowbel W., Jang B.Z. Impact behavior and impact-fatigue testing of polymer composites. *Composites Science and Technology*, **44(2)**, pp. 107-118 (1992).

[6] Aktas M., Karakuzu R., Arman Y. Compression-after impact behavior of laminated composite plates subjected to low velocity impact in high temperatures. *Composite Structures*, **89**, pp. 77–82 (2009).

[7] Bibo G.A., Leicy D., Hogg P.J., Kemp M. High-temperature damage tolerance of CRFP: Part 1: Impact characteristics. *Composites*, **25(6)**, pp. 414-424 (1994).

[8] Bibo G.A., Hogg P.J., Kemp M. High-temperature damage tolerance of CRFP: 2. Post-impact compression characteristics. *Composites*, **26(2)**, pp. 91-102 (1995).

[9] Jang B.P., Huang C.T., Hsieh C.Y., Kowbel W., Jang B.Z. Repeated impact failure of continuous fiber reinforced thermoplastic and thermoset composites. *Journal of Composite Materials*, **25(9)**, pp. 1171-1203 (1991).

[10] Ishikawa T., Sugimoto S., Matsushima M., Hayashi Y. Some experimental findings in compression after-impact tests of CF/PEEK (APC-2) and conventional CF/Epoxy flat plates. *Composites Science and Technology*, **55**, pp. 349-363 (1995).

[11] Davies P., Riou L., Mazeas F., Warnier P.. Thermoplastic composite cylinders for underwater applications. *Journal of Thermoplastic Composite Materials*, **18(5)**, pp. 417-443 (2005).

[12] Schrauwen B., Peijs T. Influence of matrix ductility and fibre architecture on the repeated impact response of glass-fibre-reinforced laminated composites. *Applied Composite Materials*, **9(6)**, pp. 331-352 (2002).

[13] Kim J-K., Sham M-L. Impact and delamination failure of woven-fabric composites. *Composites Science and Technology*, **60**, pp. 745-761 (2000).

[14] Bibo G.A., Hogg P.J. Review: the role of reinforcement architecture on impact damage mechanisms and post-impact compression behavior. *Journal of Material Sciences*, **31**, pp. 1115-37 (1996).

[15] Ghasemi Nejhad M.N., Parvizi-Majidi A. Impact behaviour and damage tolerance of woven carbon fibre-reinforced TP composites. *Composites*, **21(2)**, pp. 155-168 (1990).

[16] Wang H., Vu-Khanh T. Damage extension in carbon fiber/PEEK cross ply laminates under low velocity impact. *Journal of Composite Materials*, **28(8)**, pp. 684-707 (1994).

[17] Gao S-L., Kim J-K. Cooling rate influence in carbon fibre/PEEK composites. Part III: impact damage performance. *Composite Part A*, **32(6)**, pp. 775-785 (2001).

[18] Aymerich F., Priolo P., Vacca D. Static loading and low-velocity impact characterization of graphite/PEEK Laminates, in “Proceedings of the *International Conference on Advanced Materials*”, Hurghada, Egypt, (1998).

[19] Vedula M., Koczak M.J. Impact resistance of cross-plied polyphenylene sulfide composites. *Journal of Thermoplastic Composite Materials*, **2(3)**, pp. 154-163 (1989).

[20] Spamer G.T., Brink N.O. Investigation of the CAI properties of C/PPS and C/APC-2 thermoplastic materials in “ Proceedings of the *33rd International SAMPE Symposium: Materials-Pathway to the Future”*, pp. 284-295 (1988).

[21] Ma C-C. M., Lee C-L., Chang M-J., Tai N-H. Effect of physical aging on the toughness of CFR PEEK and PPS composites. *Polymer Composites*, **13(6)**, pp. 441-447 (1992).

[22] Vieille B., Aucher J., Taleb L. Influence de la température sur le comportement mécanique de stratifiés tissés thermoplastique ou thermodurcissable. *Revue des Composites et des Matériaux Avancés*, **21(3)**, pp. 317-343 (2011).