

THE ROLE OF PARTICLE-TOUGHENING IN IMPROVING POST-IMPACT COMPRESSIVE STRENGTH

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Abstract

This work investigates the micromechanisms associated with particle-toughening strategies to improve the damage resistant and damage tolerant performance in carbon fibre reinforced polymer (CFRP) materials. Five material systems were studied; consisting of one untoughened and four particle-toughened systems. Synchrotron radiation computed tomography (SRCT) was used to study the damage micromechanisms in standard 150mm x 100 mm rectangular coupons subjected to 30 J low velocity impact loading. Laboratory based micro-focus computed tomography (μ CT) enabled damage assessments and comparison of coupons subjected to low velocity impact, quasi-static indentation and at the onset of compression-after-impact failure. Mechanisms leading to damage resistance and damage tolerance are discussed along with strategies to use these observations to inform finite element models.

1. Introduction

Composite materials have seen a recent increase in use in the aerospace industry due to their high strength and stiffness to weight ratios. One significant remaining issue, however, is their intrinsically poor impact damage resistance, which has subsequent implications on the residual compressive strength. This is of particular concern in the barely visible impact damage (BVID) regime where impact can induce significant internal damage which is not clearly visible on the surface. Such damage can go undetected during routine in-service inspections, leading to sub-optimal design of the aircraft structure to accommodate the anticipated loss in strength. The aerospace industry, therefore, demands materials with better impact damage resistant and post-impact damage tolerant properties [1, 2]. One current strategy is to use toughening-particles dispersed within the resin at the laminate ply interfaces to increase composite toughness [3]. This has been demonstrated to work well at suppressing the propagation of delaminations; a damage mode widely reported to contribute significantly to a loss in compressive strength. Such delaminations are attributed to the formation of sub-laminates which have lower buckling stability [4, 5].

The increase in toughness has been brought about through a range of energy absorption and crack-tip shielding processes [6] which include crack-deflection, crack-bridging, crack-tip blunting, particle-matrix interface debonding and particle-induced localised yielding [6-12].

Whilst these mechanisms have been reported, it is unclear as to the relative contributions of these mechanisms significantly towards the overall toughness. Given the many parameters such as particle size, particle stiffness, particle geometry, particle volume fraction, and particle/matrix interfacial strength that can affect the damage mechanisms and toughness performance [6, 12-14], a better understanding of the contribution of each of these mechanisms towards toughness is required to guide the choice of constituents and processing.

Regarding compression after impact experiments, whilst it is generally agreed that the size of the projected damage area (an indication of the delamination area) scales with a loss in compressive strength, the mechanisms contributing to compressive failure are rather unclear [15-20]. Whilst there are numerous published models to predict the failure load of these experiments [21, 22], there is very little experimental work capturing the mechanisms involved. Experimental understanding is therefore necessary to ensure such models accurately predict the failure mechanisms. One of the key questions revolves around whether or not toughening particles offer additional gain once the coupon has been impacted, or if the local buckling failure is simply linked to the size of the damage area.

The purpose of this paper is to understand better the role particles play towards improving impact damage resistance and post-impact residual compressive strength. The study compares four particle-toughened systems and one system with no particles and focuses on identifying the key mechanisms contributing to toughness, comparisons between loading rates (low velocity impact and quasi-static indentation) and the mechanisms contributing towards a loss in compression-after-impact (CAI) strength. Based on the results obtained in this work and previously published work [23-25], mechanisms leading to damage resistance and damage tolerance are discussed along with strategies to complement these observations with finite element models.

2. Materials and test methods

2.1. Materials

Five proprietary unidirectional prepreg materials were utilised in this study. These materials consisted of one untoughened epoxy system containing no toughening particles (UT) and four particle toughened systems (T1-T4) ranked in order of impact damage resistance as measured by the size of the projected delamination area obtained through ultrasonic C-scan; T1 being the least damage resistant and T4 the most. The particle toughened systems consisted of thermoplastic particles introduced into the base epoxy resin. The difference between the particle toughened systems was the particle size and particle chemistry which was varied whilst maintaining the same particle-to-resin concentration by weight, fibre volume fraction and intermediate modulus fibre type.

For each system, ASTM D7136M standard panels were manufactured, consisting of a 24 ply layup with a [45/0/-45/90]3S stacking sequence. Panels were vacuum-bagged and fully cured under pressure in an autoclave oven to the manufacturer's specifications. Panel thickness was approximately 4.5 mm +/- 0.2 mm across the systems tested. Panels were cut to create test coupons measuring 100 × 150 mm to within the tolerances of D7136M.

2.2. Impact, quasi-static indentation and compression after impact testing

Impact testing was conducted according to the ASTM D7136M procedure at target impact energies of 25, 30, 40 and 50 J; these were repeated three times. After impact, ultrasonic C-scan was performed to measure the extent of the projected damage area.

Quasi-static indentation (QSI) testing was performed using the same boundary conditions as listed in ASTM D7136M. Out-of-plane loading was applied to the centre of the coupons using a universal testing machine at a rate of 2 mm per minute. Tests were interrupted when the out-of-plane displacement reached 2, 2.5, 3, 4 and 5 mm; this procedure was repeated three times for each material system. After each interruption, C-scan and μ CT was performed

After coupons were subjected to 25, 30, 40 and 50 J of impact, CAI testing was performed to measure the failure strength. This was conducted according to ASTM D7137M standards. To monitor damage growth at near failure loads, coupons subjected to 25 and 30 J of impact for the UT and toughened coupons respectively were μ CT scanned after impact, at the point of compressive failure and after failure.

3. Results

3.1. Impact damage resistance

Results from impact tests are plotted in Figure 1 illustrating the extent of the damage area for a given impact energy for each of the material systems. The T4 system outperformed the UT system by a factor of four, thereby demonstrating superior impact damage resistance. The T1-T3 systems show an intermediate level of impact damage resistance. The wide range of damage resistance performance between systems enabled characterisation of the mechanisms and correlation with the impact damage resistance ranking.

In previously published work [25], SRCT was performed capturing delaminations near the crack tip. One of the key observations made in the particle toughened systems, with the exceptions of T2, was the presence of particle resin debonding occurring ahead of the crack tip leading to delaminations forming within the interlaminar region. Within the T2 system, this mechanism was not observed, with the majority of delaminations occurring in the intralaminar region approximately one fibre into the ply. This difference is highlighted in Figure 2. This observation highlights the competing mechanisms involved. The lack of particle-resin debonding in the T2 system suggests that for the purposes of increasing toughness an upper limit exists to the interfacial strength, beyond which fibre-resin debonding occurs rather than confinement of the crack in the interlaminar region. This results in the crack not interacting with the particles, and thereby reducing the effective toughness. This contributes to a poorer damage resistance in comparison with the T4 system.

Regarding the particle-containing systems T1, T3 and T4, similarities between their damage micromechanisms were observed and are shown in a schematic in Figure 3. The damage consisted of particle-resin debonding occurring ahead of the crack tip, crack deflection around the particles, and bridging ligament formation. The ligament formation consisted of particles and uncracked resin bridging the crack faces. Towards the wake of the crack, particles were observed to fully debond from the resin, along with fracture of uncracked resin sites.

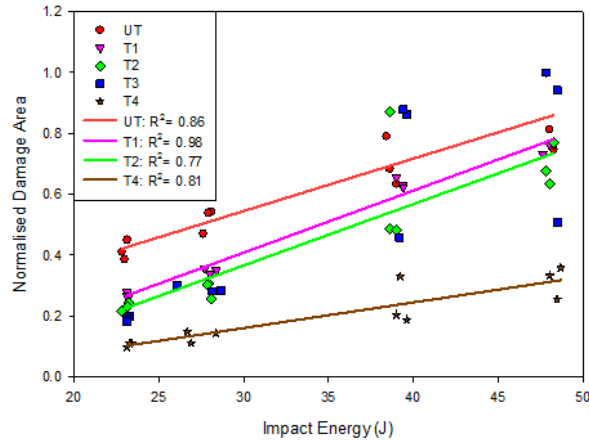


Figure 1. Plot of damage area against impact energy for each material system.

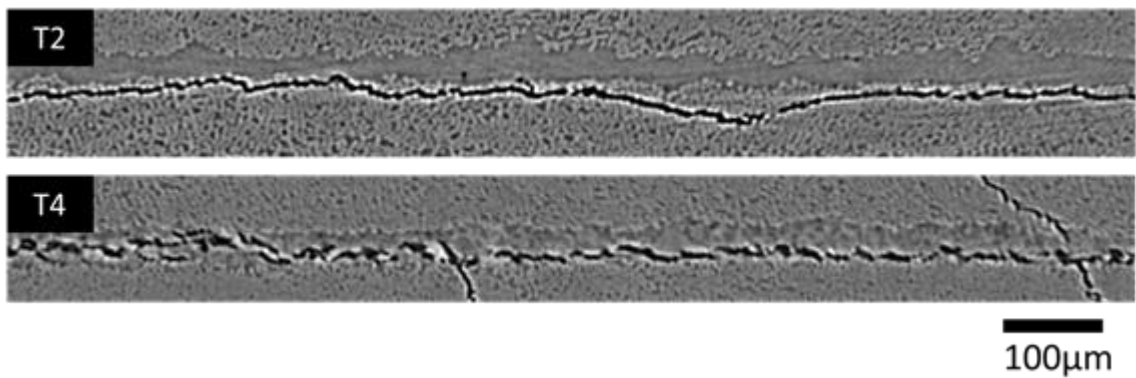


Figure 2. SRCT cross-sections of delaminations in T2 and T4 system.

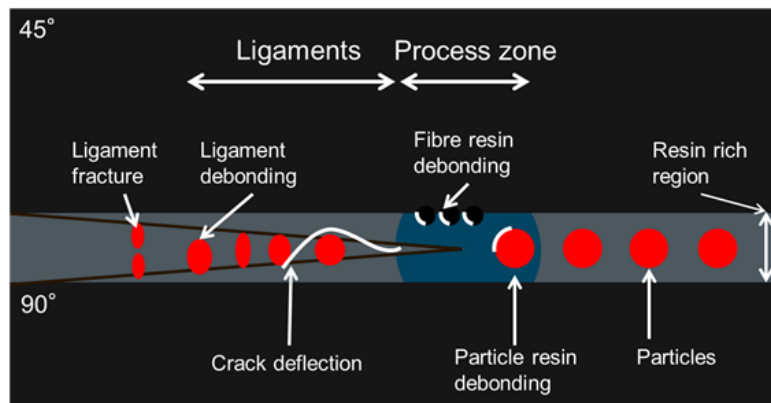


Figure 3. Schematic illustrating the particle-toughening process.

To understand the contribution of crack deflection, increase in crack path length and the extent of bridging ligament formation towards impact damage resistance, previous work was undertaken to quantify this behaviour [25]. Across the systems tested it was quantitatively found that there was marginal increases in crack path length with toughened systems suggesting that increase in fracture surface area contributed little towards toughness. Additionally, the extent of crack deflection measured by the fracture surface roughness showed that whilst particle systems do observe a rougher fracture surface, this does not correlate strongly to the impact damage resistance ranking. Regarding the extent of bridging near the crack tip, there was a correlation with the damage resistance ranking of the material systems. However, the magnitude of bridging did not correlate to same magnitude of

improvement to damage resistance. This suggests that bridging may act in conjunction with other mechanisms not measurable through μ CT. This may include, but is not limited to, plastic deformation of the bridging ligaments, the stiffness of the particles bridging the delamination to reduce crack tip stresses and the energy absorbed by particle-resin interfacial debonding. To appreciate which of these mechanisms contribute towards toughness, implementation of these parameters into microscale models are required. This will also need to take into account the competing mechanisms of fibre-resin and particle-resin debonding.

3.2. Rate dependency

In tests between impact and QSI, some differences were observed in the T1 and T3 systems regarding the increase in damage area as a function of the applied energy. This is illustrated in Figure 4, in which the T4 system showed good correlation between the two loading conditions, and the T1 system shows divergence above 30 J as circled in the plot. The increase in impact damage area above 30 J suggests a rate-dependency in this system. The differences are quite clear when observing the mechanisms under impact and QSI at similar applied energies (~ 27 - ~ 28 J respectively), as shown in Figure 5. A more extensive ligamented formation is observed under QSI loading conditions supporting the notion that bridging ligaments are important for improving damage resistance. This also highlights the rate-dependency of this micro-mechanism in certain particle-toughened systems; an issue that needs to be taken into consideration in finite element models and material development.

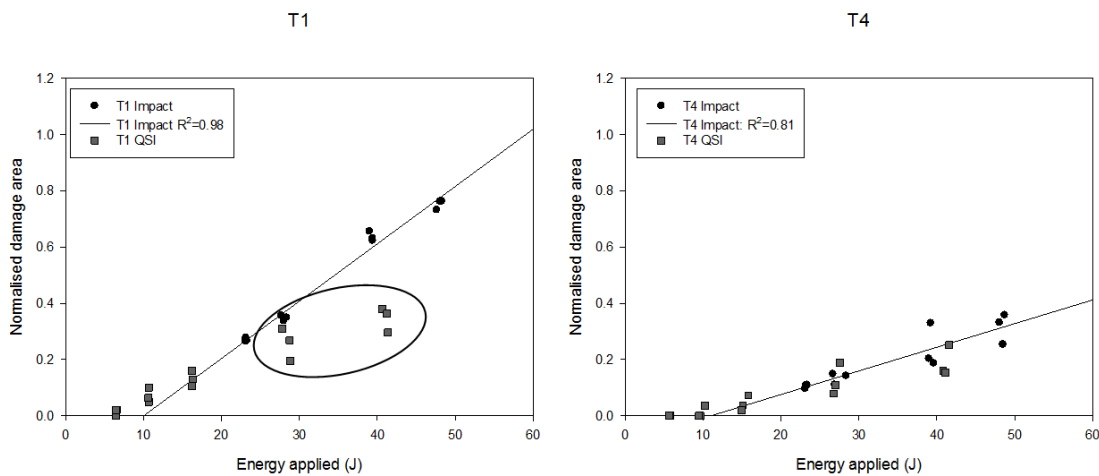


Figure 4. A comparison of damage area against energy applied for impact and quasi-static loading conditions.

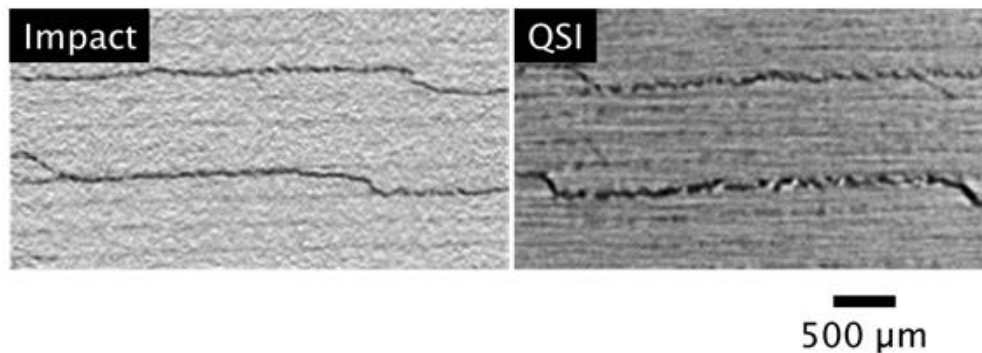


Figure 5. μ CT cross-sections of T1 coupon subjected to impact and QSI loading at the same applied energy. Note the more ligamented delamination formation under QSI loading conditions.

3.2 Compression after impact

Coupons subjected to low velocity impact were loaded in compression to measure the failure stress. This was plotted against the measured damage area to understand if this is the key parameter controlling subsequent compressive failure, see

Figure 6. Whilst the failure stress correlates with a decrease in damage area for a given material system, comparisons between particle and untoughened systems show an improvement to the failure stress for a given damage area. This was approximately 30% in the T2 and T3 systems and up to 10% improvement in the T1 system. This is interesting and suggests that particles are playing a role post-impact towards improving the residual compressive strength rather than failure stress being governed solely by the damage area created by the impact or QSI loading..

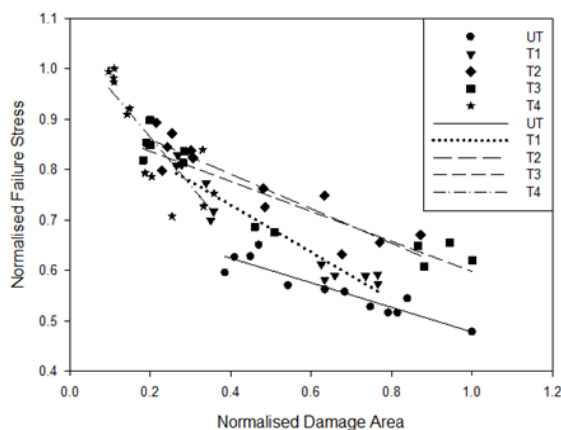


Figure 6. Plot of failure stress against damage area.

The progression of damage growth prior to CAI failure was captured as shown in Figure 7 for the UT system. Delamination growth was observed to occur into the undamaged “cone” of material lying under the impact loading point (i-ii). This was observed across all the material systems tested. No delamination growth was observed beyond the outer perimeter of the damage area prior to CAI failure. The consequence of delamination growth through the undamaged cone leads to the removal of a constraint, by unpinning the surrounding delaminations, and thereby imparting a loss in sublaminates stability, as shown in (iii) when the coupon failed. As such, the undamaged cone acts to bridge the sublaminates offering stability prior to delamination propagation in this region.

In particle-toughened systems, it is possible that bridging ligament formation could increase buckling stability of the sublaminates by providing through-thickness tractions to adjacent plies; as has been demonstrated via Z-pinning strategies [26, 27]. Additionally, the particles were seen to suppress delamination growth into the undamaged cone, in particular in the T4 system, for which very little delamination growth was observed prior to failure. The suppression of delamination growth into the undamaged cone was seen to increase sublaminates stability by enabling the undamaged cone to fully bridge this region, increasing the overall unsupported area of the sublaminates. This phenomenon could explain the ~30% improvement to failure load observed in two of the particle-toughened systems in comparison to the untoughened system.

The observations made in this study highlight important mechanisms to include in models for predicting CAI failure. In this case, delamination growth into the undamaged cone was

observed and is considered a potential controlling mechanism towards CAI failure. Additionally, bridging ligaments are likely to act to stabilise the sublaminates should they extend significantly in the wake of the crack. To confirm that these mechanisms play a significant role in determining damage tolerance, finite element models will be used to explore their influence on residual compressive strength.

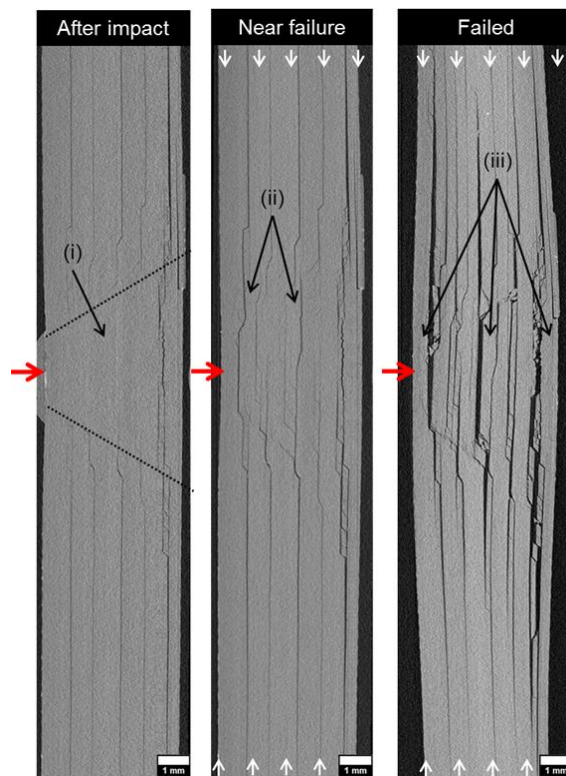


Figure 7. Progression of CAI failure showing a cross-section of a UT coupon after impact, after application of a near compressive failure load and after critical compressive failure.

4. Conclusions

Particle toughening is an important area of development for composite material systems, demonstrating a fourfold improvement in impact damage resistance for the systems examined in this study. Based on quantification of SRCT data, the extent of crack deflection and the increase in fracture surface area was shown to have little effect on the overall impact damage resistance. Regarding the extent of bridging at the crack tip, this was shown to correlate with the damage resistance of the material systems; however, the relationship was not linear, *i.e.* higher levels of bridging resulted in a decreasing rate of increase in damage resistance. It is suggested that other mechanisms not quantifiable using SRCT, operating in conjunction with bridging may be involved in contributing towards toughness. It was also observed that competing mechanisms of particle/resin debonding against fibre/resin debonding and rate-dependency play important roles in determining damage resistance. These effects require further investigation, including via modelling. In CAI experiments, particle-toughened systems improved compressive failure stress by up to ~30% for a given size of damage area. The role of particles was seen to suppress delamination growth into the post-impact undamaged “cone”; this region plays a key role in increasing the stability of the sublaminates created by the impact event. Additionally, bridging ligament formation may add stability to the sublaminates. Modelling of this phenomenon is required, however, to confirm the significance of these mechanisms.

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References

- [1] S. Abrate. Impact on composite structures: Cambridge university press; 2005.
- [2] M. Richardson and M. Wisheart, "Review of low-velocity impact properties of composite materials", *Composites Part A: Applied Science and Manufacturing*, Vol. 27, No. 12, pp 1123-31, 1996.
- [3] R.A. Pearson and A.F. Yee, "Toughening mechanisms in thermoplastic-modified epoxies: 1. Modification using poly (phenylene oxide)", *Polymer*, Vol. 34, No. 17, pp 3658-70, 1993.
- [4] S. Sánchez-Sáez, E. Barbero, R. Zaera and C. Navarro, "Compression after impact of thin composite laminates", *Composites Science and Technology*, Vol. 65, No. 13, pp 1911-9, 2005.
- [5] F. Habib, "A new method for evaluating the residual compression strength of composites after impact", *Composite structures*, Vol. 53, No. 3, pp 309-16, 2001.
- [6] S.-Y. Fu, X.-Q. Feng, B. Lauke and Y.-W. Mai, "Effects of particle size, particle/matrix interface adhesion and particle loading on mechanical properties of particulate-polymer composites", *Composites Part B: Engineering*, Vol. 39, No. 6, pp 933-61, 2008.
- [7] M. Groleau, Y.-B. Shi, A. Yee, J. Bertram, H. Sue and P. Yang, "Mode II fracture of composites interlayered with nylon particles", *Composites Science and Technology*, Vol. 56, No. 11, pp 1223-40, 1996.
- [8] J. Spanoudakis and R. Young, "Crack propagation in a glass particle-filled epoxy resin", *Journal of Materials Science*, Vol. 19, No. 2, pp 473-86, 1984.
- [9] C.H. Hsueh, "Effects of aspect ratios of ellipsoidal inclusions on elastic stress transfer of ceramic composites", *Journal of the American Ceramic Society*, Vol. 72, No. 2, pp 344-7, 1989.
- [10] F. Gao, G. Jiao, Z. Lu and R. Ning, "Mode II delamination and damage resistance of carbon/epoxy composite laminates interleaved with thermoplastic particles", *Journal of composite materials*, Vol. 41, No. 1, pp 111-23, 2007.
- [11] J.K. Shang and R. Ritchie, "Crack bridging by uncracked ligaments during fatigue-crack growth in SiC-reinforced aluminum-alloy composites", *Metallurgical Transactions A*, Vol. 20, No. 5, pp 897-908, 1989.
- [12] A. Pearson Raymond. Toughening Epoxies Using Rigid Thermoplastic Particles. Toughened Plastics I: American Chemical Society; 1993. p. 405-25.
- [13] A.C. Garg and Y.-W. Mai, "Failure mechanisms in toughened epoxy resins—A review", *Composites Science and Technology*, Vol. 31, No. 3, pp 179-223, 1988.
- [14] G.B. Chai and S. Zhu, "A review of low-velocity impact on sandwich structures", *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials Design and Applications*, Vol. 225, No. 4, pp 207-30, 2011.
- [15] L. Reis and M. De Freitas, "Damage growth analysis of low velocity impacted composite panels", *Composite structures*, Vol. 38, No. 1, pp 509-15, 1997.
- [16] M. Wisnom, "The role of delamination in failure of fibre-reinforced composites", *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, Vol. 370, No. 1965, pp 1850-70, 2012.
- [17] H. Chai, W. Knauss and C. Babcock, "Observation of damage growth in compressively loaded laminates", *Experimental Mechanics*, Vol. 23, No. 3, pp 329-37, 1983.
- [18] H. Yan, C. Oskay, A. Krishnan and L.R. Xu, "Compression-after-impact response of woven fiber-reinforced composites", *Composites Science and Technology*, Vol. 70, No. 14, pp 2128-36, 2010.
- [19] C. Soutis and P. Curtis, "Prediction of the post-impact compressive strength of CFRP laminated composites", *Composites Science and Technology*, Vol. 56, No. 6, pp 677-84, 1996.
- [20] M. Pavier and M. Clarke, "Experimental techniques for the investigation of the effects of impact damage on carbon-fibre composites", *Composites Science and Technology*, Vol. 55, No. 2, pp 157-69, 1995.
- [21] H. Suemasu, W. Sasaki, T. Ishikawa and Y. Aoki, "A numerical study on compressive behavior of composite plates with multiple circular delaminations considering delamination propagation", *Composites Science and Technology*, Vol. 68, No. 12, pp 2562-7, 2008.
- [22] A.T. Rhead, R. Butler and N. Baker, "Analysis and compression testing of laminates optimised for damage tolerance", *Applied Composite Materials*, Vol. 18, No. 1, pp 85-100, 2011.
- [23] D. Bull, L. Helfen, I. Sinclair, S. Spearing and T. Baumbach, "A comparison of multi-scale 3D X-ray tomographic inspection techniques for assessing carbon fibre composite impact damage", *Composites Science and Technology*, Vol. 75, No., pp 55-61, 2013.
- [24] D. Bull, S. Spearing, I. Sinclair and L. Helfen, "Three-dimensional assessment of low velocity impact damage in particle toughened composite laminates using micro-focus X-ray computed tomography and synchrotron radiation laminography", *Composites Part A: Applied Science and Manufacturing*, Vol. 52, No., pp 62-9, 2013.
- [25] D. Bull, A. Scott, S. Spearing and I. Sinclair, "The influence of toughening-particles in CFRPs on low velocity impact damage resistance performance", *Composites Part A: Applied Science and Manufacturing*, Vol. 58, No., pp 47-55, 2014.
- [26] X. Zhang, L. Hounslow and M. Grassi, "Improvement of low-velocity impact and compression-after-impact performance by z-fibre pinning", *Composites Science and Technology*, Vol. 66, No. 15, pp 2785-94, 2006.
- [27] A. Mouritz, "Review of z-pinned composite laminates", *Composites Part A: Applied Science and Manufacturing*, Vol. 38, No. 12, pp 2383-97, 2007.