

1 **Through-thickness crack growth resistance in fibre composites and its role in preventing ply cracking**  
2 **in cross-ply laminates**

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10 **Abstract**

11 A new mechanism is proposed to elucidate recent experimental observations of a transition from slow,  
12 stable through-thickness cracking to unstable growth in the 90° ply of a cross-ply laminate as the ply  
13 thickness increases above 40 µm for typical carbon fibre reinforced polymer composites. Herein we have  
14 identified that the transition is attributed to a rising crack-growth resistance (or R-curve) of transverse  
15 matrix cracks with increasing size. This new explanation is substantiated by obtaining the R-curve using a  
16 high-fidelity micromechanical model, followed by employing fracture mechanics principles to predict the  
17 progression and stability of through-thickness microcracking in a ply. The benefit of this new approach is  
18 that only one simulation is required to generate the R-curve, which can then be employed to predict the  
19 crack-growth behaviour for any ply thickness, instead of requiring separate simulations for each ply  
20 thickness, thereby reducing the computational burden considerably. This is particularly valuable for  
21 parametric studies to investigate the dependence on various material properties and computationally  
22 efficient analysis of large-scale structures. As illustrative examples, the dependence on matrix toughness  
23 and on volume fraction was investigated and simple linear relationships were identified for the steady-state  
24 value of crack-growth resistance. Such relationships can further reduce the computational burden,  
25 particularly when the relevant material properties may not be available from direct measurement, but can  
26 be reasonably estimated, as for cryogenic applications.

27 **Keywords:** A. Thin-ply laminate; B. Matrix cracking; C. Multi-scale modelling; C. Computational  
28 mechanics

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## 31 1. Introduction

32 Ply cracking, where transverse matrix cracks extend through the full thickness of a ply, is often the  
33 most critical initial failure mode in fibre composite laminates. This form of cracking is a major concern for  
34 composite vessels used for storing liquid or gaseous fuels, such as hydrogen and oxygen, because ply cracks  
35 can link up to form interconnected channels that cause fuel leaks and degrade the structural safety of the  
36 vessels. The pioneering work of Bailey and co-workers [1-3], as discussed in recent reviews [4, 5],  
37 identified two régimes for the thickness dependence of the ply cracking strain, or, equivalently, of the *in-*  
38 *situ* ply strength [6]. For “thin” plies, the ply cracking strain was found (experimentally and theoretically)  
39 to decrease with increasing the ply thickness, whereas for “thick” plies, the ply cracking strain is  
40 independent of the ply thickness. The transition from thin to thick régime was found to occur at a ply  
41 thickness of around 0.5 mm in cross-ply laminates, for both glass fibres (GFRP) and carbon fibres (CFRP).

42 Recent research on matrix cracking of thin-ply composites, however, has highlighted a gap in our  
43 understanding of the growth behaviour of through-thickness matrix cracks in plies of varying thickness. A  
44 through-thickness matrix crack is defined by the coalescence of at least two adjacent debondings between  
45 the fibre and matrix [7]. Slow and stable through-thickness matrix cracking was observed in laminate  
46 consisting of thin-ply of 40  $\mu\text{m}$  in thickness, whereas unstable growth was found to occur in laminates  
47 with ply thickness of 80  $\mu\text{m}$  or 160  $\mu\text{m}$ , as reported by Saito et al.[7]. These experimental observations  
48 have been closely reproduced in a high-fidelity micromechanical model by Arteiro et al. [8], whose work  
49 incorporates several recent developments in modelling of composites, relative to earlier micromechanical  
50 models [9-13], including an improved constitutive modelling for the matrix phase [14, 15]; see [16-20] for  
51 related recent work and reviews of multi-scale analysis of composites. These recent findings follow the  
52 development of tow-spreading technology, leading to commercially available ply thicknesses down to  
53 15  $\mu\text{m}$  [21, 22]. Ultra-thin plies offer benefits both because of the ability to use a larger number of ply  
54 orientations, thereby expanding the laminate design space for achieving an optimal lay-up, and because of  
55 improvements in mechanical properties, e.g. a 10% increase in the ultimate strength of quasi-isotropic  
56 CFRP laminates constructed from 40  $\mu\text{m}$  plies [23], as well as improved notched tensile strength and  
57 fatigue resistance [21]. These benefits are of particular interest for cryogenic applications such as linerless  
58 propellant tanks for reusable launch vehicles and fuel storage for deep space explorations, where resistance  
59 to ply cracking due to residual thermal stresses, thermal-mechanical cycling, and associated anti-leakage  
60 properties are crucially required [24-26].

61 The stable through-thickness growth at very small ply thickness, transitioning to unstable growth  
62 with increasing ply (or layer) thickness as revealed by experimental observations and computational model

63 predictions are not accounted for by the currently available theoretical models that tend to focus on the  
64 tunnelling mode of a pre-existing full-thickness matrix crack [27-36], whose direction of growth is parallel  
65 to fibres. This lack of fundamental understanding of the mechanism of ply-thickness effect in thin- and  
66 ultra-thin ply laminates impedes improvement in laminate design for demanding applications such as  
67 linerless cryogenic fuel tanks.

68 The present work proposes a novel proposition to account for the experimental and computational  
69 results in [7, 8] in terms of an increasing crack-growth resistance for through-thickness matrix cracks. The  
70 principle is illustrated in Fig. 1. The implementation of this principle in the present work combines the  
71 micromechanical modelling approach of Arteiro et al. [8] with the embedded cell approach [11] to derive  
72 the required crack growth resistance curve (the R-curve) in a computationally efficient manner. Next, the  
73 crack-growth behaviour is predicted by applying linear elastic fracture mechanics (LEFM) [37, 38] to  
74 determine whether crack growth is stable or unstable; details are presented in Section 4. It is worth noting  
75 that this R-curve approach was not foreshadowed in previous micromechanical simulations [8-20], or  
76 theoretical models [27-36]. One advantage of the proposed crack-growth resistance approach is that it  
77 provides a computationally efficient method for determining the critical ply thickness for the transition  
78 from stable to unstable crack growth, because the R-curve can be generated, once and for all, from a single  
79 micromechanical model, instead of the several separate models that would be required for different ply  
80 thicknesses in a trial-and-error approach. A second advantage, from the viewpoint of materials design, is  
81 that the effect of changing material properties (in particular the strength and toughness of both the matrix  
82 and/or the fibre-matrix interface) on the transitional ply thickness can be determined efficiently, simply by  
83 first calculating the resulting change in the R-curve. Similarly, the effect of operating at cryogenic  
84 temperatures can be efficiently assessed if the relevant material properties at those temperatures are known  
85 from direct measurements or can be reasonably estimated.

86 The presentation of this paper is organized as follows. The key features of the micromechanical  
87 model for a constrained layer are summarized in Section 2 using an explicit integration technique, as in our  
88 previous works [39, 40]. Representative results obtained by this approach are compared with the results  
89 reported previously in [8] that were obtained using an implicit algorithm, to demonstrate their equivalence,  
90 and to motivate the R-curve interpretation. The R-curve is then derived in Section 3, based on  
91 micromechanical modelling only within an embedded cell. In Section 4, this R-curve is employed in  
92 conjunction with finite element (FE) calculations of the energy release rate to predict the stability of  
93 through-thickness matrix cracking, according to the principle indicated in Fig. 1. Next, the dependence of  
94 the R-curve on matrix toughness and on fibre volume fraction is investigated, revealing a simple linear

95 relationship for the steady-state value. The effects of thermal residual stresses, which are ignored for  
96 simplicity in Sections 2-4, are investigated in Section 5. Finally, in Section 6, the R-curve approach is  
97 discussed relative to existing analytical models of ply cracking in laminates [27-36], highlighting the new  
98 insights provided by this approach.

## 99 **2. Micromechanical modelling of through-thickness matrix cracking in a cross-ply laminate**

100 An FE model of a cross-ply laminate is constructed that consists of three main parts, as shown in  
101 Fig. 2: (i) an inner layer incorporating a micromechanical model of one or more  $90^\circ$  plies; (ii) two adjacent  
102 constraining layers that are modelled as homogenized layers, and (iii) the interfaces between the inner layer  
103 and the outer constraining layers. The pertinent modelling assumptions are summarized in the sub-sections.

### 104 **2.1 Micromechanical model of $90^\circ$ inner layer**

105 The formulation of the micromechanical model for the inner layer follows the approach developed  
106 originally by Melro et al. [15] for an unconstrained  $90^\circ$  layer (fibres are orthogonal to the main applied  
107 stress), i.e., a periodic representative volume element (RVE), and subsequently used by Arteiro et al. [8,  
108 16] for cross-ply laminates. There are three constituents: fibres, matrix, and fibre-matrix interfaces; the  
109 relevant material properties are summarized below.

#### 110 **2.1.1 Carbon fibres**

111 The fibres are assumed to have a circular cross-section, with a diameter of  $5 \mu m$ , and to be linearly  
112 elastic and transversely isotropic relative to the fibre axis. The fibre volume fraction  $V_f$  is set at 0.6, which  
113 can be adjusted as discussed in Section 3.2. Within the  $90^\circ$  layer, the fibres are arranged in a random  
114 distribution that is representative of those observed in practice, following the approach in [15]; the  
115 importance of employing a random, rather than a regular (e.g., hexagonal) array for correct simulation of  
116 failure initiation, as well as strategies for generating representative random arrays, have been discussed by  
117 several authors [41-43]. The fibre axis is chosen to be the x-axis; the y-axis is in the direction of the in-  
118 plane loading along the  $0^\circ$  plies that is applied to cause through-thickness matrix cracking, and the z-axis  
119 is in the thickness direction, as indicated in Fig. 2(a). The relevant material properties are listed in Table 1,  
120 based on [8, 44, 45] which is representative of the IMS60 carbon fibres used in the experimental work [7].

#### 121 **2.1.2 Epoxy matrix**

122 The matrix phase is assumed to be isotropic and to conform to the elasto-plastic with damage  
123 constitutive model recently formulated by Melro et al. [14, 15]. This model involves a linear elastic régime  
124 bounded by a pressure-dependent yield criterion, and subsequent parabolic hardening that correctly  
125 captures the detailed experimental measurements of Fielder et al. [46] for a representative epoxy under

126 various forms of loading. Damage in the epoxy matrix is modelled by a single damage variable that affects  
127 only the Young's modulus once activated. Damage onset is defined by a damage activation function ( $\Phi_m^d$ )  
128 similar to the yield criterion, but with the tensile and compressive yield strengths replaced by ultimate  
129 strengths. The Bazant-Oh [47] crack band model is employed to mitigate mesh-size dependency following  
130 damage localization due to strain softening. The relevant material properties to implement this constitutive  
131 model are listed in Table 2, which are considered here as standard values for typical Bisphenol-A type plain  
132 epoxy resin as used in [7]. Further details of implementation are presented in the Supplementary Material  
133 and in [8, 14].

### 134 *2.1.3 Fibre-matrix interfaces*

135 The fibre-matrix interface is characterized by the cohesive zone model (CZM) available in Abaqus  
136 [48]. This involves a bilinear traction-displacement relation, with a high initial stiffness  $K = 10^8 \text{MPa/mm}$   
137 to enforce displacement continuity across the interface prior to damage. In the present work, damage onset  
138 is governed by the maximum stress criterion [48]. Final failure is characterized by the Benzeggagh-Kenane  
139 law [48] for mixed-mode failure. Values of the relevant properties are listed in Table 3.

### 140 *2.2 Homogenized outer constraining plies*

141 The outer layers in Fig. 2(a) are modelled as linear-elastic, orthotropic solids with the homogenized  
142 properties represent  $0^\circ$  plies, to simulate the experimental set up of Saito et al. [7]. The relevant thermo-  
143 mechanical properties are listed in Table 4, which are determined by FE homogenization of an RVE, as in  
144 [49]. The interfaces between the inner and outer layers are modelled by CZM, using representative values  
145 for interlaminar properties [8], which are listed in Table S1 in Supplementary Material.

### 146 *2.3 Model discretization, loading and boundary conditions*

147 The geometry shown in Fig. 2(a) is modelled using solid elements (C3D8R) [48] for matrix, fibres  
148 and homogenized outer layers. The average element size within the inner layer is set to be  $0.35 \mu\text{m}$ , to  
149 generate a well-structured, high-quality mesh, as suggested in [17-20], based on a sensitivity analysis. The  
150 fibre-matrix interface and the interface between inner and outer layers are modelled using cohesive  
151 elements (COH3D8) [48] of zero thickness. The model length in the loading direction ( $y$ -axis) is  $200 \mu\text{m}$ ,  
152 whereas the thickness in the fibre direction ( $x$ -axis) is approximately twice the average element size of the  
153 inner layer, as in [8]. Periodic boundary conditions are applied in the  $x$  and  $y$  directions, using linear multi-  
154 point constraints, to impose a homogenized strain,  $\varepsilon_{yy}$  that increases gradually to a final value of 2%.

155 As noted in Section 1, Abaqus/Explicit [48] is used in the present work, which allows a simpler  
156 programming of the nonlinear constitutive model for the matrix phase as a user-defined subroutine

157 VUMAT. To assess the equivalence of the explicit approach with the implicit approach employed in [8], a  
158 comparison is made for a thin-ply laminate with an inner layer of thickness  $20\ \mu\text{m}$  and outer layers of  
159 thickness  $75\ \mu\text{m}$ , employing the same material properties as [8] for model verification purposes. The results  
160 are presented as Fig. S2 in the Supplementary Material, confirming the equivalence between the two  
161 approaches.

## 162 **2.4 Simulation of through-thickness matrix cracking under increasing strain**

163 Figure 2(b) shows two representative images of the pattern of matrix cracking within an  $30\ \mu\text{m}$ -  
164 thick inner  $90^\circ$  layer, obtained in the present work. Microcracking is found to initiate at the fibre-matrix  
165 interface (i.e. debonding), at a location where the inter-fibre spacing between two neighbouring fibres that  
166 are aligned with the load axis (the  $y$ -axis) is relatively small, as indicated by a dashed box in Fig. 2(b). This  
167 is in accord with the previous experimental and computational observations [7, 8, 50]. A through-thickness  
168 matrix crack in micromechanical model is defined by the coalescence of at least two neighbouring  
169 debondings, as defined in the experimental work [7]. The progression of through-thickness matrix cracking  
170 is also similar to that previously reported in [7, 8], with a representative final pattern for an applied strain  
171 of 2% being shown in Fig. 2(b). Figure 2(c) shows in more detail the evolution of the crack opening profile  
172 for the through-thickness matrix crack indicated by the dashed box in Fig. 2(b). The localized crack opening  
173 displacement (COD) is measured as the relative displacement between adjacent nodes after the occurrence  
174 of microcracking, as in [8]. This progression of the crack opening profile under increasing applied strain  
175 values is reported here for the first time.

176 The increasing values of COD for the near tip opening profiles appear to be indicative of an  
177 increasing crack growth resistance with increasing crack length. An initial estimate of the crack growth  
178 resistance  $G^R$ , based on the tip-to-tip crack length  $2a$  of a through-thickness matrix crack and the maximum  
179 COD for various levels of strain, as obtained from Fig. 2(c), is shown in Fig. 2(d), which clearly indicates  
180 an increasing crack growth resistance. The details of the calculation are documented in the Supplementary  
181 Material. This novel interpretation of the simulation results will be explored more quantitatively in the  
182 present work by independently deriving the R-curve based on micromechanical modelling in Section 3.

## 183 **3. Crack growth resistance curve (R-curve) method**

### 184 **3.1 Micromechanical calculation of the R-curve**

185 Unlike the fracture toughness, which can be regarded as a material property under conditions of  
186 small-scale yielding [37, 38], the R-curve is not strictly a material property: it depends on the configuration  
187 (i.e., the specimen geometry, initial crack length, type of loading, etc. [51, 52]), although in practice this

188 dependence may be masked by the scatter in experimental measurements. This configuration dependence  
189 is revealed, however, by analytical models where the material property characterizing failure is specified  
190 in the form of a traction-separation law [53], and in experimental work involving large-scale bridging [51,  
191 52]. Accordingly, it is important to generate an R-curve based on a configuration that is as close as possible  
192 to the intended application of the R-curve for predictive purposes.

193 In the present context, an attractive option for generating the R-curve computationally is to use two  
194 neighbouring fibres, such as those in the neighbourhood of the initiation site in Fig. 2(b), as load application  
195 points, with equal and opposite point forces applied to those two fibres to initiate failure at the fibre-matrix  
196 interface. It was found, however, that this approach resulted in inelastic deformation of the matrix around  
197 those fibres that is not directly associated with the crack initiation and crack growth process, and the  
198 contribution of this extraneous energy dissipation to the work of fracture could not readily be quantified.  
199 These results are presented in the Supplementary Material.

200 Instead, the configuration shown in Fig. 3 was found to be more tractable for generating an  
201 appropriate R-curve. In this figure, the entire model consists of  $90^\circ$  ply material only. However, for  
202 computational efficiency, micromechanical modelling is limited to only a central region of the specimen,  
203 while the remainder is modelled with homogenized (elastic) properties, as indicated in Fig. 3, in accordance  
204 with the embedded cell approach [11]. The size of this embedded cell must be such as to fully capture all  
205 inelastic deformation. This requirement must be verified at the end of a simulation. In the present work, the  
206 height of the embedded cell in the  $y$ -direction (the direction of loading) was taken to be  $h = 76 \mu m$ , which  
207 was found to satisfy the above requirement, as will be seen below. The specimen length in the  $z$ -direction  
208 (the direction of crack growth) was taken to be  $w = 420 \mu m$ , which proved to be large enough for the R-  
209 curve to reach a plateau value  $G_{SS}^R$  corresponding to steady-state crack growth. The same material properties  
210 and element types as described in Section 2 were again employed. The displacement field is assumed to be  
211 continuous across the interfaces between the embedded cell and the homogenized outer portions. The  
212 average element size within the embedded cell is  $0.35 \mu m$  approximately, and the element size increases  
213 gradually to  $6.3 \mu m$  in the homogenized portions. As in Section 2.3, the model thickness in the  $x$ -direction  
214 (the fibre axis) is twice the average element size within the embedded cell, and periodic boundary conditions  
215 are applied to the faces normal to the  $x$ -axis.

216 To avoid the difficulties noted above with point loading of fibres, the model shown in Fig. 3 includes  
217 a pre-existing crack of length  $a_0 = 4 \mu m$ , created by initially debonding the relevant nodes. This length is  
218 chosen on the basis that it is equal to half of the shortest tip-to-tip crack length that can be reliably measured  
219 experimentally or in simulations [7, 8], as indicated in Figs. 2(b,c), and for which the crack can reasonably

220 be approximated as a straight centre crack for the purposes of calculating the energy release rate, keeping  
 221 in mind that the initial crack at the fibre-matrix interface has a curvature dictated by the fibre diameter,  
 222 which is  $5 \mu m$  for the present simulations. To generate the R-curve, this pre-crack is opened by applying  
 223 specified displacements on the face  $z = 0$ : all nodes with  $y > 0$  are given a specified vertical displacement  
 224  $u_y(y > 0, z = 0)$ , whereas all nodes below the crack, with  $y < 0$ , are held fixed, i.e.  $u_y(y < 0, z = 0) =$   
 225  $0$ . The resulting nodal forces for  $y > 0$  are summed to obtain the crack opening force  $P$ , whereas the crack  
 226 mouth opening displacement is given by  $\Delta = u_y(y > 0, z = 0) - u_y(y < 0, z = 0)$ .

227 A representative load-displacement ( $P - \Delta$ ) curve obtained in this manner is shown in Fig. 4(a).  
 228 This curve includes some periodical unloadings that are intended to check that the unloading response is  
 229 linearly elastic, with the unloading curves returning to the origin, as required to ensure the validity of the  
 230 LEFM formula [54] for calculating the crack growth resistance, viz.,  $G^R = \frac{1}{2}bP^2\partial C/\partial a$ , where  $b$  denotes the  
 231 specimen thickness (in the  $x$ -direction in the present case) and  $C = P/\Delta$  the compliance. To obtain the crack  
 232 length  $a$ , the crack tip is identified by searching for the element furthest away from the face  $z = 0$  at which  
 233 the damage variable has reached the value 1, corresponding to complete failure; the crack length is then  
 234 taken as the distance from the crack tip to the face  $z = 0$ .

235 Because of the random distribution of fibres built into the model, slightly different results are  
 236 obtained from different simulation runs. Figure 4(b) shows the combined results obtained from five  
 237 different simulations, involving five different realizations of random fibre distributions, but all for the same  
 238 volume fraction. The scatter in these results is attributable to the randomness of the fibre distributions, all  
 239 other properties being deterministic. For predictive purposes, the raw results from the simulations are fitted  
 240 with a rational polynomial of second order, to obtain the continuous curve shown in Fig. 4(b) for  $G^R(a)$ .  
 241 With the present approach, this curve necessarily starts from the assumed initial crack length  $a_0 = 4 \mu m$ .  
 242 However, the smallest ply thickness that can currently be achieved with tow-spreading technology is around  
 243  $15 \mu m$  [21, 22], so that this assumed initial crack length is not unduly restrictive in practice.

244 Figure 4(c) illustrates the progression of deformation and crack growth under increasing load. It can  
 245 be seen that the crack path often follows the fibre-matrix interface, and proceeds discontinuously, with  
 246 heavily deformed but unbroken matrix ligaments bridging the crack behind the crack tip. These  
 247 observations from the present micromechanical modelling are in accord with previous experimental and  
 248 computational results [7-20, 50]. Thus, the crack path is not straight at the microscopic level. Figure 4(d)  
 249 shows representative maps of the damage activation variable ( $\Phi_m^d$ ) for two crack lengths. It can be seen  
 250 that the damage zone is fully contained within the embedded cell, even for the longer crack length of  $80 \mu m$ ,

251 corresponding to steady-state crack growth, indicating that the selected size for the embedded cell is  
252 sufficient to correctly capture the R-curve.

### 253 **3.2 Effect of matrix toughness and fibre volume fraction on crack growth resistance**

254 The influence of various material properties on through-thickness matrix cracking can now be  
255 investigated by first determining their influence on the R-curve. To illustrate the procedure, consider first  
256 the influence of the matrix fracture toughness  $G_c$ , all other material properties and modelling assumptions  
257 being kept the same. Figure 5(a) shows the R-curves obtained for  $G_c = 80,160,240 J/m^2$ , as well as the  
258 previous curve in Fig. 4(b) for  $G_c = 120 J/m^2$  which serves as the baseline. It can be seen that the steady-  
259 state value  $G_{SS}^R$ , and the extent of crack growth required to reach the steady state, both increase with  
260 increasing  $G_c$ . Recalling that crack growth involves debonding at the fibre-matrix interface, with the crack  
261 being bridged by matrix ligaments behind the crack tip, it appears reasonable to expect that the steady-state  
262 value  $G_{SS}^R$  can be estimated from a simple rule-of-mixtures (RoM) formula involving both the interfacial  
263 toughness ( $G_{Ic}^{int}$ ) and the matrix toughness, as follows

$$264 \quad G_{SS}^R = V_f G_{Ic}^{int} + (1 - V_f) G_c. \quad (1)$$

265 Figure 5(b) shows that this simple formula indeed provides a good estimate for  $G_{SS}^R$ , at least within the  
266 investigated range for  $G_c$ . This in turn suggests a useful approach for estimating  $G_{SS}^R$  in situations where  
267 measured values of  $G_c$  may not be available but can be reasonably estimated.

268 Next, Fig. 5(c) shows the R-curves for fibre volume fractions  $V_f = 0.45$  and  $0.55$  in addition to the  
269 baseline value  $0.6$  employed previously in Fig. 4(b), and retaining the same values for all other parameters.  
270 It can be seen that the initial slope of the R-curve ( $\partial G^R / \partial a$ ) and the final steady-state value  $G_{SS}^R$  increase  
271 with decreasing  $V_f$ . Figure 5(d) shows that the simple formula in Eq. (1) again provides a reasonable  
272 estimate for  $G_{SS}^R$ , at least for the range of  $V_f$  investigated here that is representative of the values encountered  
273 in practice. The increase in  $G_{SS}^R$  is consistent with an increased dissipation due to wider bridging ligaments  
274 as  $V_f$  decreases (the fibre diameter remaining fixed).

## 275 **4. Stability of through-thickness matrix cracking**

276 Having obtained the R-curve for the crack growth resistance  $G^R(a)$  as in Fig. 4(b), the next  
277 requirement is to determine the energy release rate  $G(a)$  as a function of the half-crack length  $a$  for the  
278 configuration shown in Fig. 6(a).

279 **4.1 Energy release rate**

280 To determine  $G(a)$  the inner layer is modelled as a homogeneous layer, with the same lamina  
 281 properties as listed in Table 4 for the outer plies, but with the fibre orientation now parallel to the  $x$ -axis,  
 282 instead of the  $y$ -axis. The deformation in Fig. 6(a) is assumed to be plane strain ( $\varepsilon_{xx} = 0$ ). The energy  
 283 release rate can then be expressed as follows [29, 55]

$$284 \quad G(a) = \Lambda \sigma_{yy}^2 \pi a f \left( \frac{a}{t_1}, \frac{t_2}{t_1}, \frac{E_T}{E_L}, \dots \right) \quad (2)$$

285 where  $\Lambda = \left( \frac{1}{E_T} - \frac{\nu_L}{E_L} \right)$ ,  $\sigma_{yy} = \varepsilon_{yy} / \Lambda$ , which denotes the stress within the inner layer in an uncracked  
 286 laminate, under an applied strain  $\varepsilon_{yy}$ . In this expression, the function  $f$  can be regarded as a nondimensional  
 287 correction factor that depends on several configurational parameters and material properties. In the limit of  
 288 very small cracks, the influence of the outer ply thickness and stiffness becomes negligible, i.e.,  
 289  $f(a/t_1 \ll 1) = 1$ , and  $G(a)$  reduces to  $G(a) = \Lambda \sigma_{yy}^2 \pi a$ , as which is appropriate for a centre crack of  
 290 length  $2a$  in a transversely isotropic material under plane strain. Values of  $G(a)$  can be readily determined  
 291 computationally, e.g. by the virtual crack closure technique (VCCT) available in Abaqus [48]. The results  
 292 for the dependence of  $G(a)$  on crack size are shown in Fig. 6(b), using the following dimensionless form:

$$293 \quad g \left( \frac{a}{t_1} \right) = \frac{G(a)}{\Lambda \sigma_{yy}^2 \pi t_1} = \frac{a}{t_1} f \left( \frac{a}{t_1}, \frac{t_2}{t_1}, \frac{E_T}{E_L}, \dots \right) \quad (3)$$

294 These results were obtained for a fixed value of the constraining layer thickness  $t_2 = 480 \mu m$ , as in the  
 295 experimental work [7]. It can be seen that  $g(a/t_1)$  is only weakly dependent on the modulus ratio  $E_T/E_L$   
 296 in the range 0.02–0.1; it was also found to be insensitive to the value of the inner-layer thickness  $2t_1$  in the  
 297 range 40 – 160  $\mu m$ , and insensitive to the Poisson ratios within the range encountered in practice for  
 298 CFRP. Furthermore, the present results for orthotropic layers are very close to the results for isotropic inner  
 299 and outer layers, with Young's moduli equal to  $E_T$  and  $E_L$  respectively [29, 55]. The important feature in  
 300 Fig. 6(b) is that  $g(a/t_1)$  decreases when the crack size exceeds 70% of the inner layer thickness, which  
 301 indicates the constraining action of the stiff outer layers, as noted in previous work [29]. For the subsequent  
 302 analysis, the numerical results for  $g(a/t_1)$ , for  $E_T/E_L = 0.05$ , were fitted to a fourth-order polynomial in  
 303  $a/t_1$ . More information is given in the Supplementary Material.

304 **4.2 Predicting the progression of through-thickness matrix cracking**

305 The progression of through-thickness matrix cracking can now be predicted by the procedure  
 306 indicated in Fig. 1, which is illustrated in more detail in Fig. 6(c). Crack growth is stable if the gradient of  
 307 the crack-growth curve is less than the gradient of the crack growth resistance curve, i.e.,  $\frac{\partial G}{\partial a} < \frac{\partial G^R}{\partial a}$ , where

308  $G, G^R, a$  denote respectively the crack-growth energy release rate, crack-growth resistance, and crack length  
 309 (or half-crack length for centre cracks). Unstable crack growth occurs when the gradient of the crack-  
 310 growth curve exceeds the gradient of the crack growth resistance curve, i.e.,  $\frac{\partial G}{\partial a} > \frac{\partial G^R}{\partial a}$ .

311 Consider first the case where the inner layer thickness is  $40 \mu m$ , corresponding to a single ply in  
 312 the experimental set up, i.e.  $n = 1$  for the  $[0_m, 90_{n/2}]_s$  laminates investigated by Saito et al. [7]. The curves  
 313 of  $G(a)$  for that inner-layer thickness  $2t_1$  are shown in red in Fig. 6(c), for various values of  $\varepsilon_{yy}$ . It can be  
 314 seen that the crack growth criterion  $G = G^R$  is first satisfied when  $\varepsilon_{yy} = 0.7\%$ , and the half-crack length  
 315  $a_0 = 4 \mu m$ . This initial value of crack length  $a_0$  corresponds to the shortest crack length, corresponding  
 316 one fibre-matrix disbond, for which the R-curve is available, as discussed above in Section 3. It is clear  
 317 from Fig. 6(c) that for this initial crack length,  $\partial G/\partial a < \partial G^R/\partial a$ , thus crack growth is predicted to be  
 318 stable [37, 38]. Next, as the applied strain increases to  $\varepsilon_{yy} = 1\%$ , the crack grows to a length of  $a = 7 \mu m$   
 319 and  $\partial G/\partial a < \partial G^R/\partial a$ , i.e., crack growth is still predicted to be stable. Proceeding in this manner, one can  
 320 construct a continuous curve of crack length versus applied strain, which is shown as the solid red curve in  
 321 Fig. 7(a). The crack length is normalized by the layer thickness to facilitate comparison with the  
 322 experimental results in [7] which are shown as data points in Fig. 7. It can be seen that the R-curve method  
 323 correctly captures the average behaviour observed experimentally, and correlates well with the results  
 324 obtained in the present work by the micromechanical simulation described in Section 2, which are shown  
 325 by the dashed curve in Fig. 7. Stable crack growth is always predicted until full through-thickness  
 326 penetration at  $\varepsilon_{yy} = 1.7\%$ .

327 Consider next an inner layer thickness  $2t_1 = 160 \mu m$ , corresponding to  $n = 4$  for the experimental  
 328 laminate [7]. It can be seen from Fig. 6(c) that the crack growth criterion is again satisfied at  $\varepsilon_{yy} = 0.7\%$   
 329 for  $a_0 = 4 \mu m$ , and growth is predicted to be initially stable as  $\partial G/\partial a < \partial G^R/\partial a$ . However, when the  
 330 applied strain has increased to  $\varepsilon_{yy} = 1\%$ , and the half-crack length has reached  $a = 11 \mu m$ , further crack  
 331 growth is predicted to be unstable, because now  $\partial G/\partial a > \partial G^R/\partial a$ . The full predicted response is shown  
 332 by the solid blue curve in Fig. 7(b), which can again be seen to correctly capture the behaviour recorded  
 333 experimentally, as well as the results obtained by the micromechanical simulations described in Section 2.

334 Finally, for the intermediate layer thickness investigated experimentally,  $2t_1 = 80 \mu m, n = 2$ , the  
 335 predicted behaviour is slightly more complicated. Crack growth is again predicted to initiate at  $\varepsilon_{yy} = 0.7\%$   
 336 for  $a_0 = 4 \mu m$ , and to be initially stable because  $\partial G/\partial a < \partial G^R/\partial a$ , as can be seen from the green curves  
 337 in Fig. 6(c). Next, when the applied strain has increased to  $\varepsilon_{yy} = 1\%$ , and the crack length has reached

338  $a = 11 \mu m$ , subsequent growth is predicted to be unstable, because  $\partial G/\partial a > \partial G^R/\partial a$ . But now this  
339 inequality only applies for a limited extent of crack growth: as the crack grows, the value of  $G$  eventually  
340 decreases and intersects the  $G^R$  curve for  $a = 32 \mu m$  ( $a/t_1 = 0.8$ ), and  $\partial G/\partial a < \partial G^R/\partial a$  at that point,  
341 which indicates crack arrest. Further crack growth beyond that point requires an increasing applied strain.  
342 This subsequent growth is predicted to be stable, because  $\partial G/\partial a < 0$  beyond  $a/t_1 \approx 0.7$ , whereas  
343  $\partial G^R/\partial a \geq 0$ , so that the stability criterion  $\partial G/\partial a < \partial G^R/\partial a$  is always satisfied until full through-  
344 thickness penetration at  $\varepsilon_{yy} = 1.4\%$ . This predicted response is shown by the continuous green curve in  
345 Fig. 7(b), and it can be seen to be in reasonable agreement with the experimental observations in [7], as  
346 well as the results of micromechanical simulations of the present work.

## 347 **5. Through-thickness matrix cracking at cryogenic temperature**

348 Thermal residual stresses arise at two length scales in fibre-composite laminates [4, 56]: intra-ply  
349 residual stresses arise between the fibres and matrix within a ply, due to their different coefficients of  
350 thermal expansion (CTE); inter-ply residual stresses arise between plies of differing orientations within a  
351 laminate, due to the variation in the homogenized CTE with orientation. The inter-ply residual stresses are  
352 uniformly distributed across the ply thickness, and can be calculated by using conventional lamination  
353 theory [57] or FE, whereas the intra-ply residual stresses require micromechanical modelling. To account  
354 for the effects of thermal residual stresses on the R-curve approach for predicting through-thickness crack  
355 growth, two distinct calculations are required: first, the change in the R-curve must be determined, based  
356 on micromechanical modelling, and secondly, the change in the energy release rate as a function of applied  
357 strain  $\varepsilon_{app}$  must be evaluated, by first calculating the inter-ply residual stress or strain  $\varepsilon_{th}$ , based on  
358 lamination theory. Furthermore, the change in material properties with decreasing temperature must also  
359 be taken into account, most notably the change in matrix fracture toughness. These steps are addressed in  
360 turn in the following sub-sections.

### 361 **5.1 Effect of intra-ply residual stresses on the crack growth resistance curve**

362 The implementation of the FE model described in Section 3.1 is modified by including a preliminary  
363 step, henceforth referred to as Step 1, to simulate a temperature change  $\Delta T$  prior to applying an external  
364 load in Step 2, while retaining all other model specifications and material properties as previously described  
365 in Section 3. For illustrative purposes,  $\Delta T$  is chosen here to be  $-278K$ , corresponding to the temperature  
366 drop from room temperature (RT) to liquid hydrogen temperature (LH<sub>2</sub>). At the completion of Step 1, the  
367 resulting residual stress field is similar to that reported in several previous micromechanical simulations of  
368 unconstrained unidirectional plies [10, 41]. Consequently, this residual stress field will not be documented  
369 in detail here, except to note (i) the stress everywhere remains below the damage-inducing levels, both in

370 the matrix and at the fibre-matrix interface, for the current choice of  $\Delta T$  and material parameters; and (ii)  
 371 the interfacial normal stress is predominantly compressive (see Fig. S8 of the Supplementary Material),  
 372 suggesting an increased resistance to crack growth due to delayed fibre-matrix debonding.

373 The residual stress field from Step 1 is retained as an initial stress field for Step 2 of determining  
 374 the R-curve. For that purpose, an initial edge crack of length  $a_0 = 4 \mu m$  is again introduced and opened as  
 375 previously described in Section 3.1 and Fig 3. The results are shown in Fig. 8(a), for the same four values  
 376 of matrix toughness  $G_c$  that were previously used to generate the results in Fig. 5(a). The solid curves in  
 377 Fig. 8(a) again represent a rational polynomial fit to the data obtained from five separate simulations, for  
 378 each value of  $G_c$ . The R-curves previously derived without considering the thermal residual stresses are  
 379 shown as dashed curves for comparison purposes. It can be seen that the residual stresses indeed lead to an  
 380 increased crack growth resistance. Furthermore, the steady-state resistance  $G_{SS}^R$  again increases linearly with  
 381 increasing  $G_c$ , and there is an increasing difference with the previously calculated values of  $G_{SS}^R$  with  
 382 increasing  $G_c$ , as shown in Fig. 8(b), which is not captured by the RoM in Eq. (1), but Eq. (1) nevertheless  
 383 continues to provide a useful estimate for practical purposes.

## 384 **5.2 Effect of inter-ply residual stresses on the energy release rate**

385 Turning next to the calculation of the energy release rate for the configuration shown in Fig. 6(a), it  
 386 is noted that Eq. (2) is still applicable provided that  $\sigma_{yy}$  is taken as the sum of the thermal residual stress  
 387 and the applied stress, or equivalently, that  $\varepsilon_{yy}$  is interpreted as the total strain  $\varepsilon_{yy}^{total}$  on the  $90^\circ$  layer,  
 388 consisting of an residual strain at the ply level  $\varepsilon_{yy}^{th}$ , plus an applied strain  $\varepsilon_{yy}^{app}$  at LH<sub>2</sub>:

$$389 \varepsilon_{yy}^{total} = \varepsilon_{yy}^{th} + \varepsilon_{yy}^{app}. \quad (4)$$

390 The calculation for  $\varepsilon_{yy}^{th}$  is documented in the Supplementary Material.

## 391 **5.3 Predicting through-thickness crack growth at cryogenic temperature**

392 Having determined the effects of thermal residual stresses on both the R-curve and the energy  
 393 release rate, we can now predict the progression of through-thickness matrix cracking as in Section 4. The  
 394 predicted responses are shown as the solid curves in Figs. 9(a-c), for the three  $90^\circ$  layer thicknesses and  
 395 four values of  $G_c$  previously considered in Section 4. These predictions are compared with the results  
 396 obtained by modifying the micromechanical model in Section 2 to include a preliminary step (Step 1)  
 397 corresponding to a temperature change  $\Delta T = -278K$ , prior to applying an external load. These simulation  
 398 results here serve the role of experimental results, in the absence of direct experimental observations at  
 399 LH<sub>2</sub>, and they are shown as the dashed lines in Figs. 9(a-c), but for only two values of matrix toughness

400  $G_c = 120$  and  $240 \text{ J/m}^2$  due to the laborious computational effort. It can be seen that the predicted crack  
401 growth based on the R-curve approach is again in good agreement with the more laboriously obtained  
402 micromechanical simulations.

403 In Figs. 9(a-c), the dotted vertical lines indicate the thermal strain  $\varepsilon_{yy}^{th}$  on  $90^\circ$  layer at the completion  
404 of Step 1. Thus, the predicted behaviour as a function of applied strain can be visualized by focussing on  
405 the results to the right of these dotted lines. In particular, it is of interest to note the value of applied strain  
406 that is required to achieve full-thickness cracking, which is re-plotted in Fig. 9(d). For a given value of  $G_c$ ,  
407 crack growth becomes progressively more unstable as the ply thickness increases, thereby resulting in full-  
408 thickness cracking at a lower applied strain; on the other hand, for a given ply thickness, growth becomes  
409 more stable as  $G_c$  increases, so that a higher applied strain is required for full-thickness cracking. These  
410 results provide a basis for quantifying the potential benefits of the nano-toughening techniques, as  
411 developed in [58, 59], in conjunction with tow-spreading technology for facilitating the design and  
412 optimization of the composite vessels for storing cryogenic fuels.

#### 413 ***5.4 Effect of changing matrix toughness with decreasing temperature***

414 Variations in matrix and interface strength and toughness are generally very laborious to  
415 characterize experimentally [58, 59], on the one hand, but are expected to have a significant influence on  
416 the failure behaviour, on the other. To illustrate how the present work provides an efficient approach for  
417 estimating matrix-cracking behaviour at cryogenic temperatures, consider the case where the matrix  
418 toughness decreases from a RT value  $G_c = 120 \text{ J/m}^2$ , to an assumed lower value  $G_c = 60 \text{ J/m}^2$  at LH<sub>2</sub> as  
419 measured in [58] at cryogenic temperature and as indicated by the arrow in Fig 8(b) (all other material  
420 properties being assumed to retain their RT values, for simplicity). Figures 10(a-c) shows the predicted  
421 behaviour for through-thickness matrix cracking, based on (i) the RT value of  $G_c$ , and ignoring thermal  
422 residual stresses for which the total strain is simply the applied strain  $\varepsilon_{yy}^{total} = \varepsilon_{yy}^{app}$ , and (ii) the LH<sub>2</sub> value  
423 of  $G_c$ , and accounting for thermal residual stresses for which the total strain is the sum of thermal residual  
424 strain and applied strain as defined by Eq. (3). It is shown that the ply thickness for which through-thickness  
425 matrix cracking transitions from stable to unstable growth is smaller for case (ii) relative to case (i), due to  
426 the lower  $G_c$  at LH<sub>2</sub>; on the other hand, the applied strain for causing full-thickness cracking is significantly  
427 lower for case (ii) relative to case (i) (see Fig. 10(d)), due to the high inter-ply residual strain at LH<sub>2</sub>. This  
428 is clearly an important observation from the viewpoint of exploiting the potential benefits of toughened thin  
429 plies for cryogenic applications.

## 6. Discussion

### 6.1 Contribution to computational efficiency for micromechanical simulations

Micromechanical modelling is becoming an essential tool to characterize the deformation and failure mechanisms within a ply, providing insights and inputs for the next levels (mesoscale and macroscale) in a structured multiscale modelling framework leading to a virtual testing pyramid for fibre-composite structures [18]. The recent micromechanical model of Arteiro et al. [8] correctly captures the experimentally observed transition from unstable to stable through-thickness crack growth in the 90° ply of a cross-ply laminate [7]. However, the specification of material properties in [8] requires no fewer than 35 parameters, as well as two stress-strain curves characterizing the plastic deformation and ultimate strength of the bulk epoxy, as derived from the experimental measurements [46] for tension and compression. The predicted response depends to some extent on each of these input parameters, but a detailed parametric analysis is clearly prohibitive. Even a more restricted study of the dependence on matrix and interfacial strength and toughness would be daunting, particularly if one were to also include an integrated modelling of the curing response for more accurate calculation of thermal residual stresses, as recently proposed in [20]. From this perspective, the present work contains two important contributions: first, a new approach has been proposed that can significantly reduce the computational burden, especially for a systematic parametric analysis; secondly, the simulations reported in [8, 16] have been extended to include the effects of residual stresses.

The benefit of the R-curve approach in reducing the computational burden has already been noted in Sections 4,5: once the R-curve has been generated the progression of through-thickness cracking can be readily predicted for any ply thickness, instead of requiring separate simulations for each ply thickness of interest. Furthermore, to determine the influence of various material parameters on the ply failure response, it is sufficient to determine their influence on the R-curve, which again reduces the required number of separate micromechanical simulations.

### 6.2 Comparison with theoretical models of ply cracking

Figure 11 shows the ply-thickness dependence of the failure strain for full through-thickness cracking, as derived from the present micromechanical modelling and experimental observations [7], as well as the ply cracking strain predicted by current theoretical models [27-36]. It is important to understand the scope as well as the limitations of both sets of predictions. Currently available theoretical models for ply cracking are based on (i) fracture mechanics [27-29], or (ii) finite fracture mechanics [31]. In the latter approach, there is no attempt to track the progression of cracking either through the thickness, or across the width, of a ply. Instead, a failure criterion is formulated by equating the change in configurational energy

462 (i.e. the sum of the stored elastic energy and the potential energy of the loading mechanism) due to a ply  
463 crack (spanning the full thickness and width of the ply) to the work of fracture for that area of crack. This  
464 is the approach originally adopted by Bailey et al. [1-3], and subsequently refined by several others on the  
465 basis of more refined stress analyses for evaluating the energy change, notably based on variational  
466 principles [31-35]. Regardless of the level of sophistication of the stress analysis, a finite fracture mechanics  
467 approach will necessarily predict a ply cracking strain that decreases as  $1/\sqrt{2t_1}$  with increasing ply  
468 thickness  $2t_1$ , as shown by the dashed green curve in Fig. 11, and thus cannot account for the transition to  
469 the thick-ply régime, a limitation that was recognised in [2, 3].

470 Dvorak and Laws [29] have formulated a fracture mechanics approach that can account for the thin-  
471 to-thick-ply transition, as shown by the dashed red lines in Fig. 11, albeit based on postulating the existence  
472 of an effective initial crack whose precise shape and size are not known *a priori*, and employing simplified  
473 two-dimensional (2D) analyses that envisage the failure critical event as being either (i) crack growth across  
474 the ply thickness, for thick plies, or (ii) crack growth in a tunnelling mode (i.e. across the width of the  
475 laminate, for a crack spanning the full thickness of the transverse ply) for thin plies. In both cases, the  
476 relevant critical value of the energy release rate  $G_{mc}$  is assumed to be a constant, albeit having a different  
477 value for through-thickness cracking relative to crack growth across the laminate width (tunnelling mode).  
478 This approach necessarily predicts that through-thickness cracking is always unstable once it initiates  
479 (unless the pre-existing initial crack is assumed to span more than 70% of the ply thickness, cf. Fig. 6), and  
480 thus is unable to account for the experimental and computational observations of slow, stable growth in [7,  
481 8]. Furthermore,  $G_{mc}$  has the status of an empirical constant that is determined by curve fitting the  
482 experimentally measured data for failure strain versus ply thickness. Although this empirical value of  $G_{mc}$   
483 will necessarily depend on the fracture toughness for the bulk matrix, there is no explicit relation between  
484 the two, and hence no simple way of predicting the beneficial effects of matrix toughening, for example.  
485 The dashed curves in Fig. 11 are based on assuming  $G_{mc} = 220 \text{ J/m}^2$  as originally used in [29, 34]. It  
486 should be noted that the energy change driving the tunnelling mode is essentially the same as that considered  
487 in the finite fracture mechanics approach; the difference between the dashed curves in Fig. 11 can be  
488 attributed to differences in the stress analyses for calculating this energy change.

489 By contrast to the models in [27-30], the micromechanical simulations do not assume a pre-existing  
490 crack. Instead, cracking is shown to initiate by fibre-matrix debonding, as shown earlier in Fig. 2(b). This  
491 pattern of initiation and subsequent growth correlates closely with experimental observations [7, 28],  
492 whereas experimental evidence of pre-existing cracks, as required by the fracture mechanics models [27-  
493 29], has never been reported. Another difference with the present work is that the models in [27-29, 31-35]

494 have always assumed a single value for the fracture toughness, rather than a rising R-curve, as derived here  
495 in Section 3. This R-curve behaviour has been shown to account for the transition to slow, stable through-  
496 thickness crack growth with decreasing ply thickness. The predicted strain for full through-thickness  
497 cracking based on the present R-curve approach is displayed as the solid curve in Fig. 11, showing good  
498 agreement with experimental observations [7] and micromechanical simulation results. A limitation of the  
499 present micromechanical model, however, is that it can only characterize through-thickness cracking, and  
500 therefore it cannot predict the ply-cracking strain if the failure critical event is crack growth across the  
501 laminate width (tunnelling mode). However, these predictions for through-thickness cracking are most  
502 valuable for laminate design, particularly in the context of ensuring antileakage properties. Future work is  
503 required to clarify the relation between the resistance to through-thickness cracking and the so-called matrix  
504 cracking toughness that characterizes the tunnelling mode of ply cracking. The steady-state crack resistance  
505 for through-thickness cracking,  $G_{SS}^R$ , has been shown in the present work to be well approximated by a RoM  
506 formula, Eq. (1), and it is therefore necessarily less than the epoxy matrix toughness,  $G_c$ , whereas the  
507 matrix-cracking toughness  $G_{mc}$ , which determined empirically as discussed above, is generally much larger  
508 than  $G_c$ ; for example, the dashed curves in Fig. 11 employed a value of  $G_{mc}$  that is approximately twice the  
509 fracture energy for a typical epoxy matrix. The reason for this relatively large value of the matrix-cracking  
510 toughness is not entirely clear; it may be due to fibre-bridging, which cannot be capture with the present  
511 micromechanical model.

## 512 7. Conclusions

513 A new concept has been proposed and demonstrated to account for the experimental observations  
514 of slow, stable cracking across the thickness of the 90° ply in a cross-ply laminate, when the ply thickness  
515 is reduced to 40 μm. The proposed explanation is that through-thickness cracking proceeds under an  
516 increasing crack growth resistance characterized by an R-curve, after initiating in a natural manner at the  
517 fibre-matrix interface. This explanation has been substantiated by employing a high-fidelity  
518 micromechanical model to generate the R-curve for crack growth in an unconstrained 90° ply. The R-curve  
519 is then employed to predict the progression and stability of through-thickness cracking based on principles  
520 of fracture mechanics. The predicted crack growth correctly captures both the experimental observations  
521 and the simulation results, showing a transition from stable growth to unstable growth with increasing 90°-  
522 layer thickness.

523 One major benefit of the R-curve approach is that only one micromechanical simulation is required  
524 for generating the R-curve, which can then be employed to predict the progression of crack growth for inner  
525 layers of any thickness, instead of requiring a separate simulation for each layer thickness of interest. This

526 considerably reduces the computational burden for micromechanical simulations, which is particularly  
527 valuable for conducting detailed parametric studies to examine the dependence of ply cracking on various  
528 material properties. As an illustrative example, the dependence on matrix toughness has been investigated.  
529 The results show that the steady-state value of crack growth resistance appears to increase linearly with  
530 increasing matrix toughness, and this linear increase is closely approximated by a simple rule-of-mixtures  
531 formula that also involves the interfacial toughness and fibre volume fraction.

532 This first demonstration of the R-curve approach can be extended to account for the effects of  
533 residual stresses, which arise at two distinct length scales. The intra-ply residual stresses must be evaluated  
534 using micromechanical modelling, and they only affect the R-curve, whereas the inter-ply residual stresses  
535 must be evaluated from ply-level (mesoscale) modelling, and they only affect the energy release rate for  
536 through-thickness cracking. It has been shown that the steady-state crack growth resistance again follows  
537 a linear increase with increasing matrix toughness. The existence and identification of such simple  
538 relationships can further reduce the computational burden for systematic parametric studies and sensitivity  
539 analyses based on micromechanical simulations. The new insights provided by these simulations relative  
540 to currently available theoretical models for ply cracking have been discussed.

#### 541 **Declaration of competing interest**

542 The authors declare that they have no known competing financial interests or personal relationships that  
543 could have appeared to influence the work reported in this paper.

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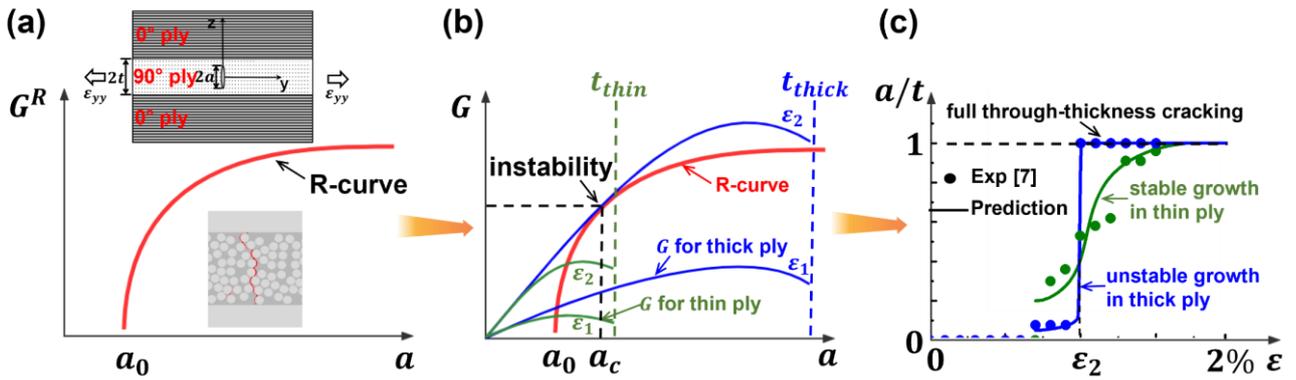
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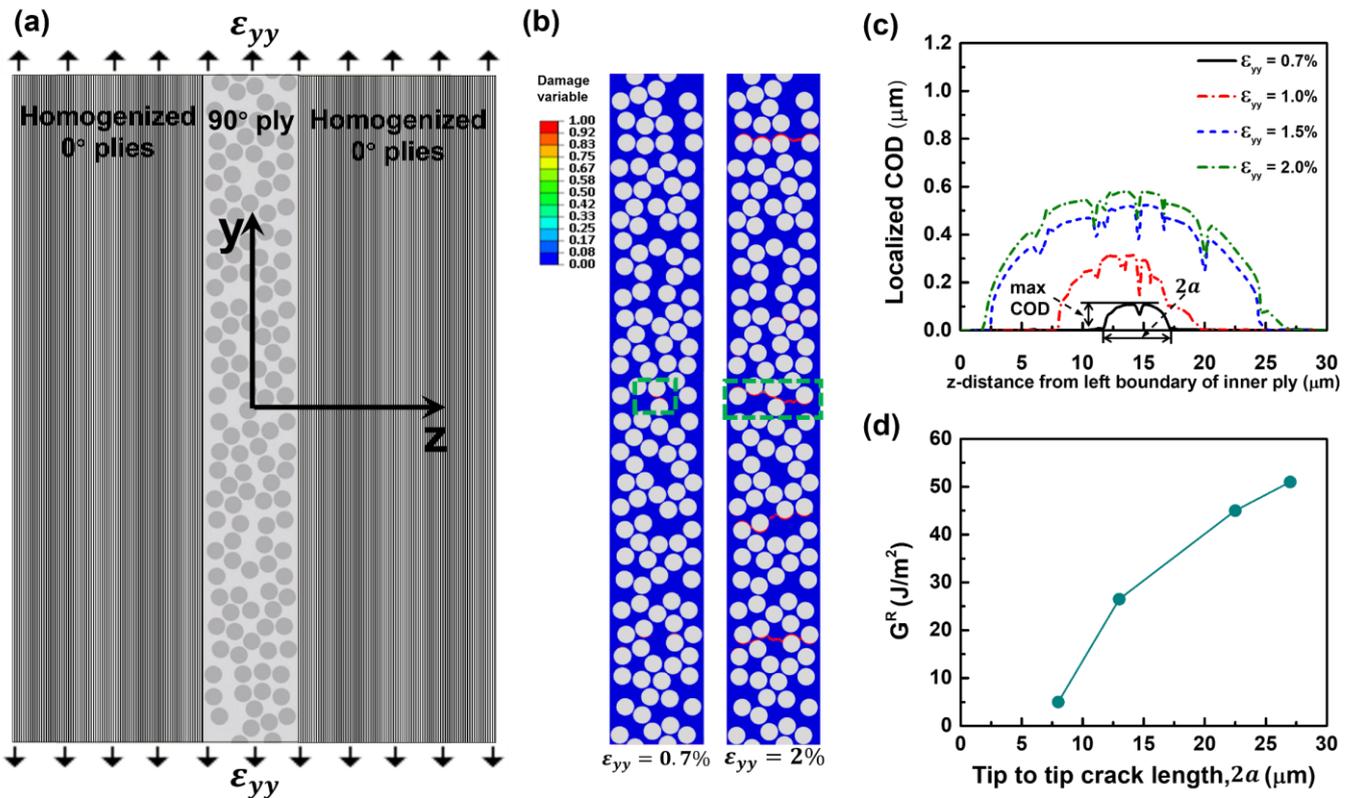
**Fig. 1** Predicting the stability of crack growth. (a) A schematic R-curve (inset shows a centre crack in a 90° ply); (b) variation of the energy release rate with crack length for a thin ply (green curves) and a thick ply (blue curves), for two values of applied strain; (c) predicted crack growth behaviour.

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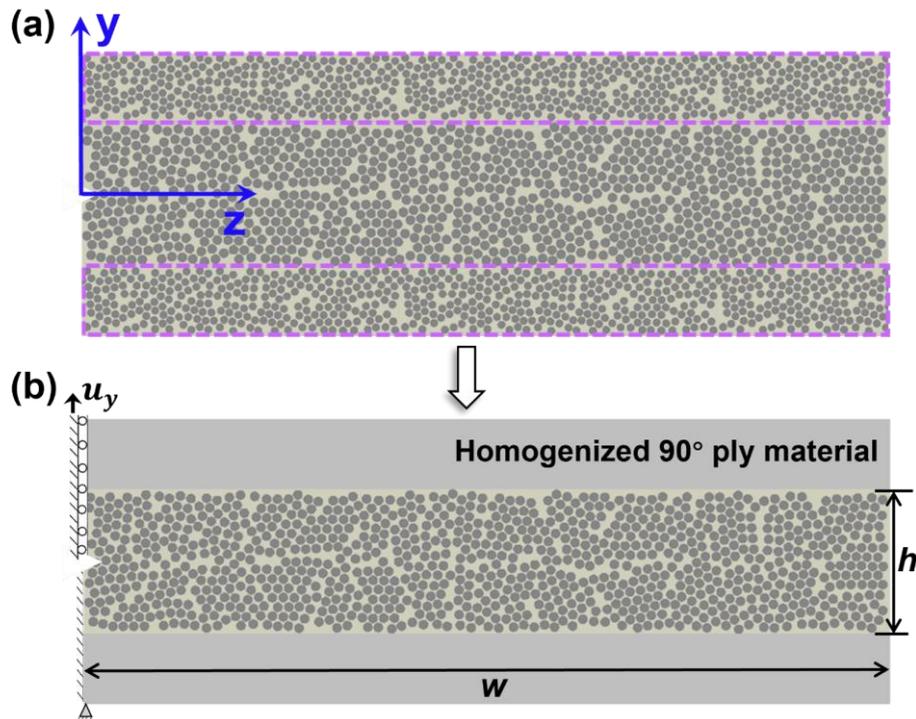
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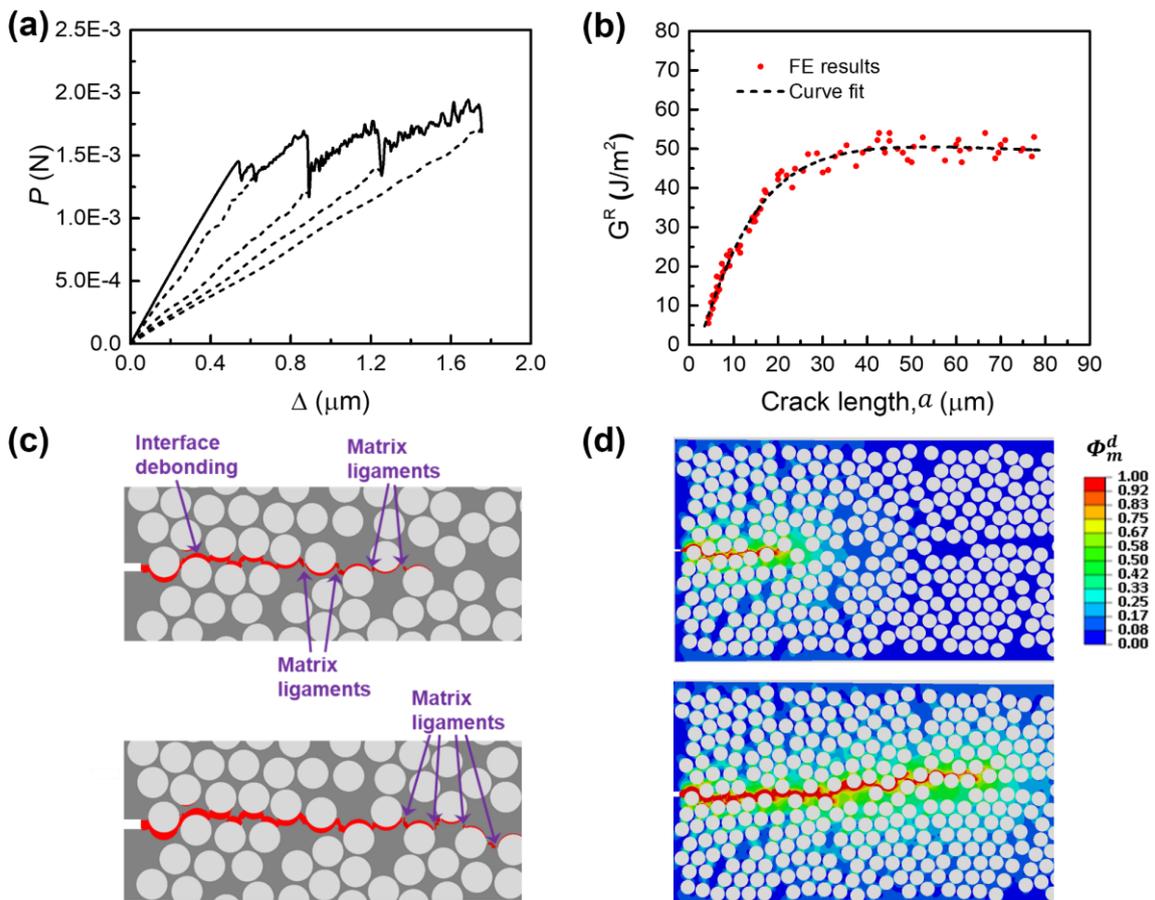


**Fig. 2** Micromechanical simulation of through-thickness cracking in a crossply laminate. (a) Model geometry; (b) simulation results for two values of applied strain, with the location of failure initiation indicated by a dashed box; (c) crack opening profiles for increasing applied strain; (d) crack growth resistance as estimated from (c).



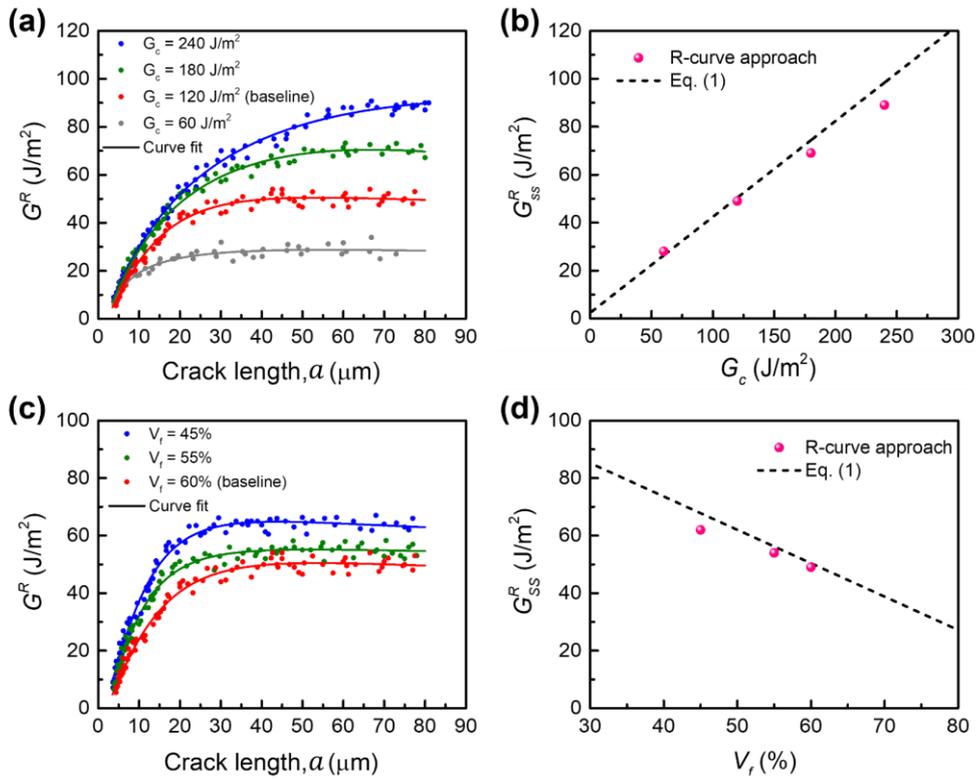
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655 **Fig. 3** Micromechanical simulation of the R-curve. (a) Cross-sectional view of 90° ply showing the coordinate  
 656 axes; (b) detailed micromechanical modelling only within an embedded cell, for computational efficiency.



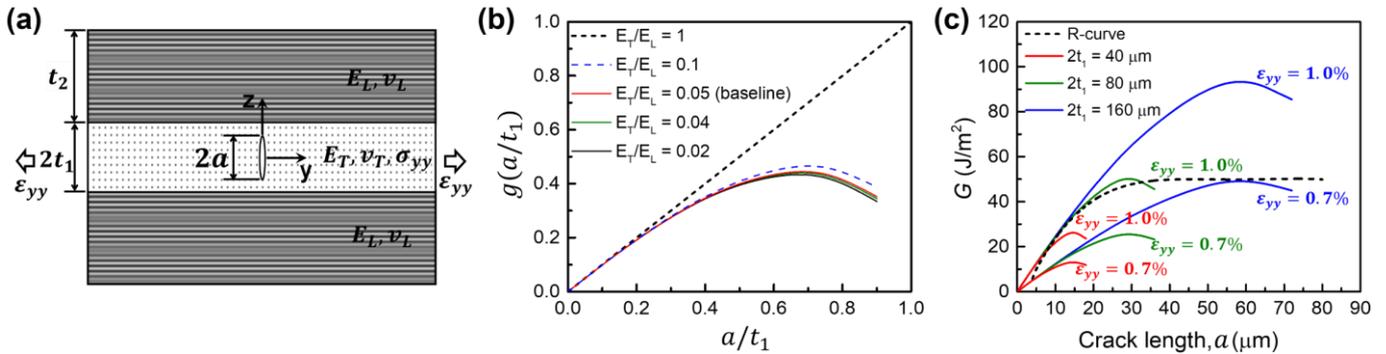
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658 **Fig. 4** The R-curve. (a) A representative load-displacement plot; (b) R-curve derived from the simulation results;  
 659 (c) crack path showing bridging by unbroken ligaments behind the advancing crack tip; (d) contour map of the  
 660 damage activation variable showing that damage zone is well contained within the embedded cell.



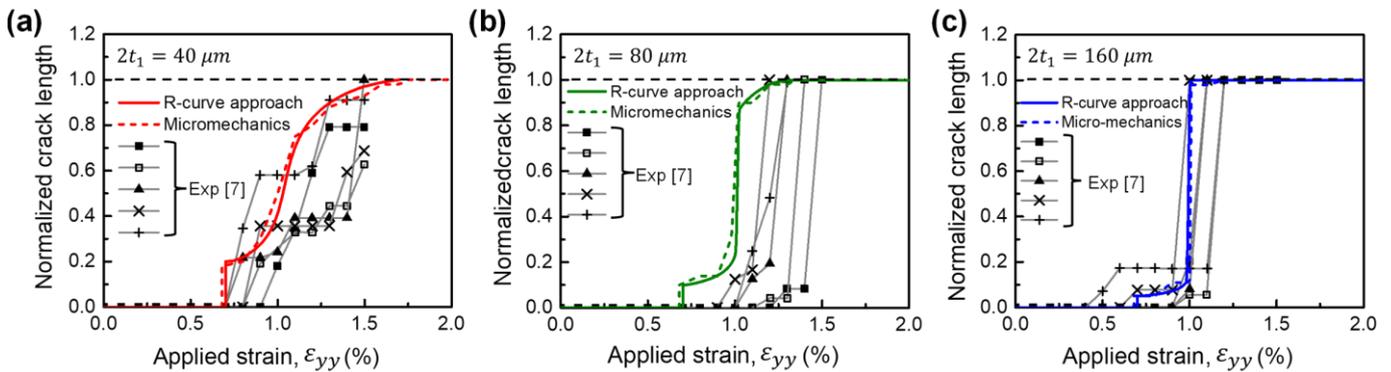
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662 **Fig. 5** Parametric variations of the R-curve. (a) R-curves for various values of matrix toughness; (b) linear  
 663 variation of the steady-state resistance  $G_{SS}^R$  versus matrix toughness, compared with a rule-of-mixtures estimate  
 664 (dashed line); R-curves for various values of fibre volume fraction; (d) linear variation of  $G_{SS}^R$ , compared with  
 665 rule-of-mixtures estimate (dashed line).



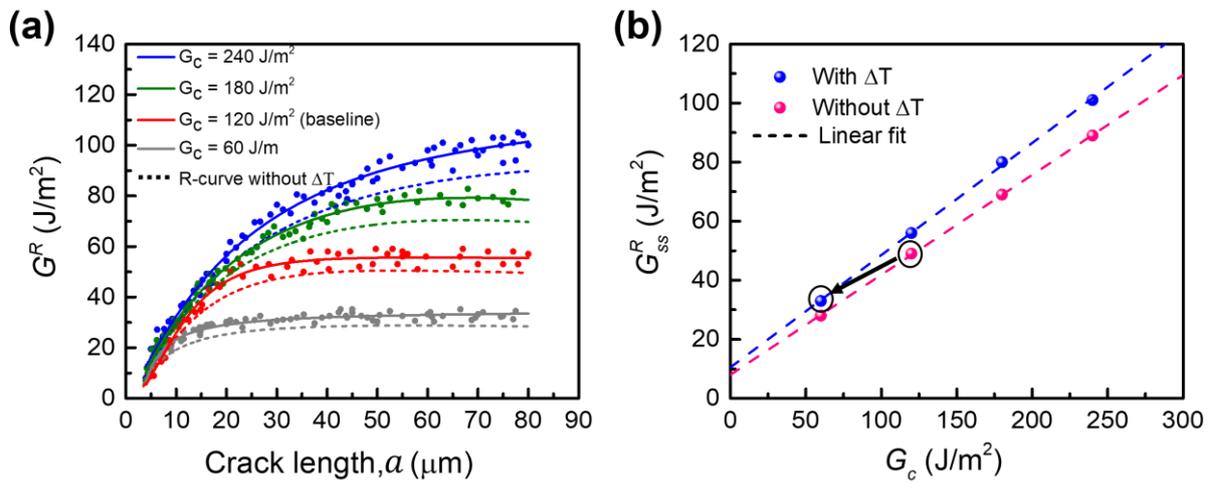
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667 **Fig. 6** Energy release rate for through-thickness cracking. (a) Model geometry and applied load; (b) normalised  
 668 energy release rate vs crack length for various ply thicknesses, and two values of applied strain.



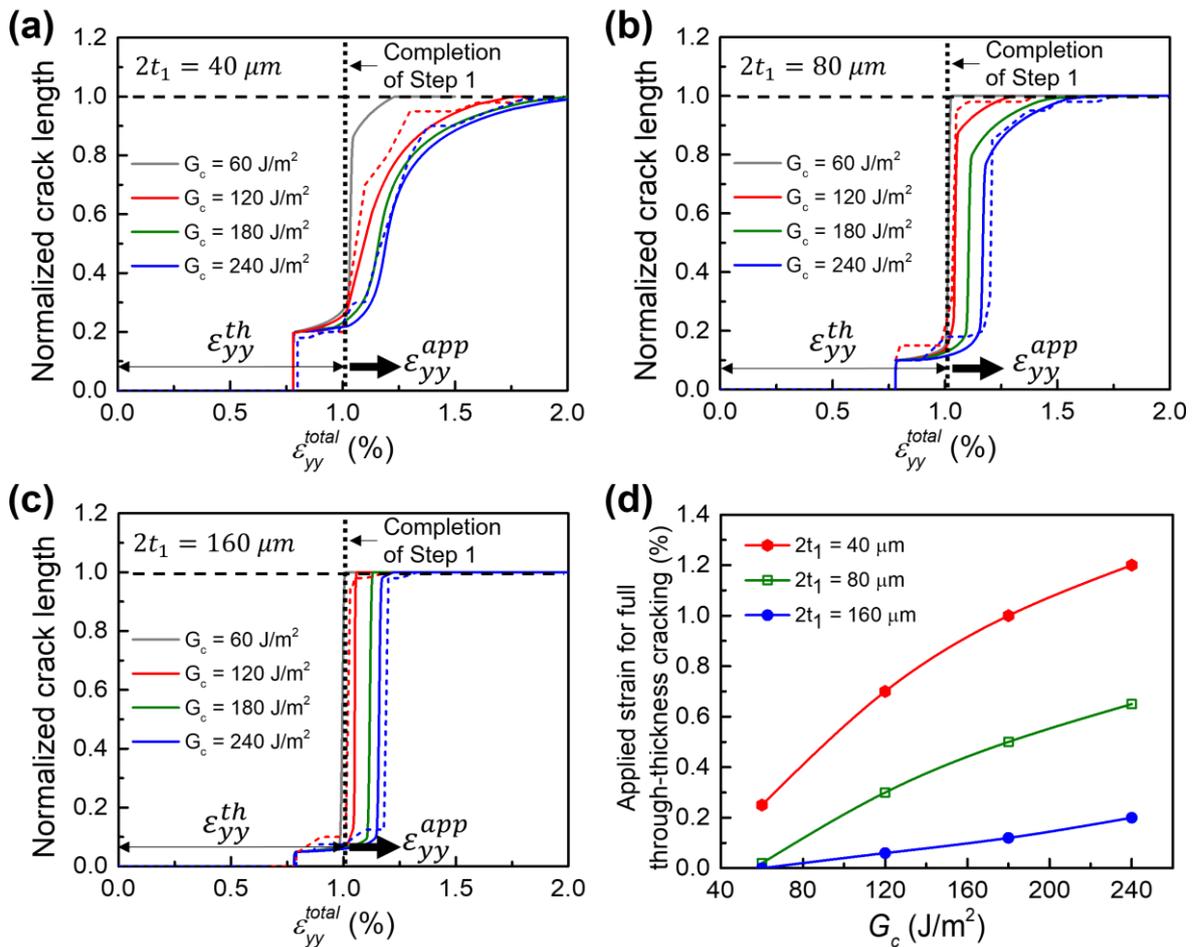
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670 **Fig. 7** Predicted crack growth behaviour based on R-curve (continuous curves), compared with the experimental  
 671 observations in [7], shown as data points, and micromechanical simulation results (dashed curves). (a) Inner  
 672 layer thickness  $2t_1 = 40 \mu\text{m}$ ; (b)  $2t_1 = 80 \mu\text{m}$ ; (c)  $2t_1 = 160 \mu\text{m}$ .



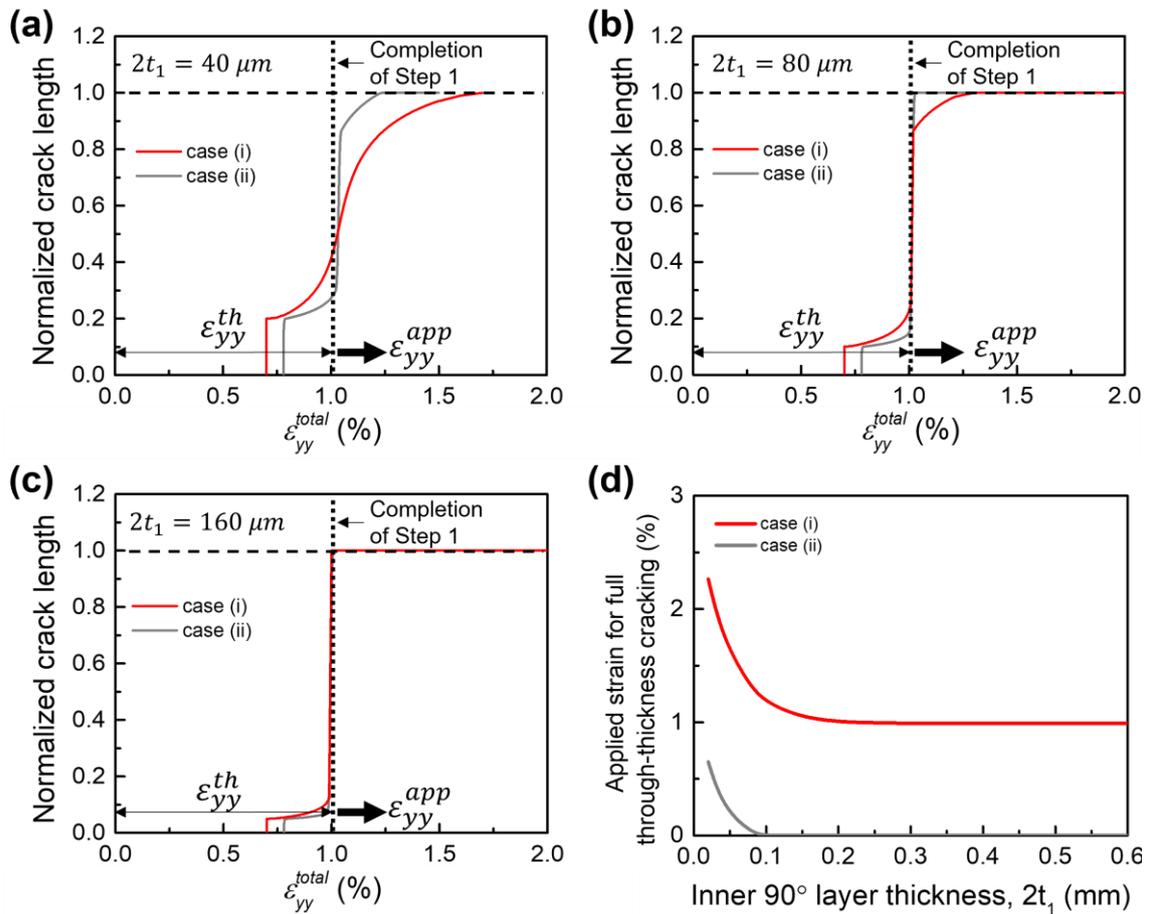
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674 **Fig. 8** R-curves in the presence of intra-ply thermal residual stress. (a) Increased crack growth resistance relative  
 675 to simulations without residual stress; (b) linear variation of steady-state resistance  $G_{ss}^R$  versus matrix toughness;  
 676 the arrow indicates the predicted change in  $G_{ss}^R$  if the matrix toughness is changed from its RT value to its LH<sub>2</sub>  
 677 value.



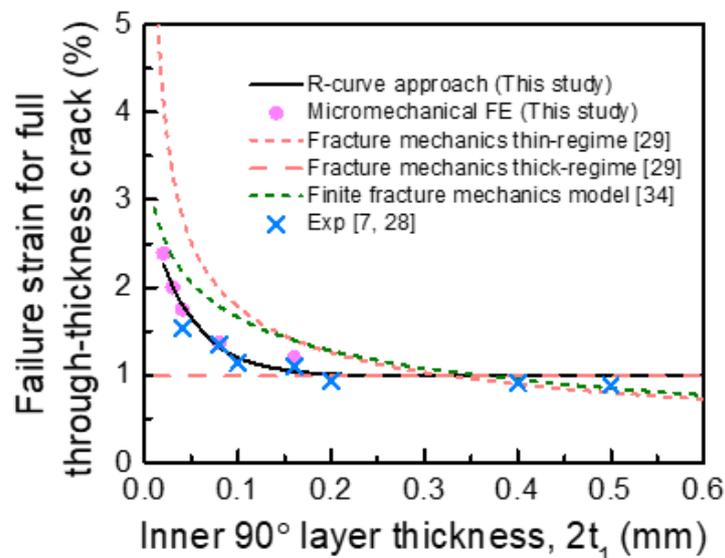
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679 **Fig. 9** Predicted crack growth behaviour as a function of total strain (i.e. thermal and applied strain), for various  
 680 ply thicknesses. (a) Inner layer thickness  $2t_1 = 40 \mu\text{m}$ ; (b)  $2t_1 = 80 \mu\text{m}$ ; (c)  $2t_1 = 160 \mu\text{m}$ ; (d) applied strain for  
 681 full through-thickness cracking versus matrix toughness.



682

683 **Fig. 10** Predicted crack growth behaviour considering a reduction in matrix toughness at low temperature. (a)  
 684 Inner layer thickness  $2t_1 = 40 \mu\text{m}$ ; (b)  $2t_1 = 80 \mu\text{m}$ ; (c)  $2t_1 = 160 \mu\text{m}$ ; (d) applied strain for full through-thickness  
 685 cracking versus  $90^\circ$ -layer thickness.



686

687 **Fig. 11** Comparison of ply failure strain for full through-thickness cracking based on the present R-curve  
 688 approach with experimental observations and with currently available theoretical models for ply cracking.  
 689 (Note: readers are referred to [29, 34] for the implementation of both analytical models. The constant  $G_{mc} =$   
 690  $220 \text{ J/m}^2$  as originally used in [29, 34] is retained here, and no thermal residual stress is considered for  
 691 simplicity.)

**Table 1**

Carbon fibre material properties

Young's moduli		Poisson's ratio	Shear moduli		Coefficient of thermal expansion		Density
$E_L$ (MPa)	$E_T$ (MPa)	$\nu_L$	$G_L$ (MPa)	$G_T$ (MPa)	$\alpha_L$ ( $K^{-1}$ )	$\alpha_T$ ( $K^{-1}$ )	$\rho$ ( $kg/mm^3$ )
279000	15000	0.2	15000	7000	$-0.7 \times 10^{-6}$	$12 \times 10^{-6}$	$1.79 \times 10^{-9}$

692 Note: the subscript  $L$  and  $T$  denote the longitudinal and transverse, respectively.**Table 2**

Epoxy matrix material properties

Material property	Value	Ref.
Young's modulus		
$E$ (MPa)	3760	[14]
Poisson's ratio		
$\nu$	0.39	[46]
Plastic Poisson's ratio		
$\nu_p$	0.3	[8]
Fracture toughness		
$G_c$ ( $J/m^2$ )	120	[58]
Tensile strength		
$X_m^t$ (MPa)	93	[46]
Compressive strength		
$X_m^c$ (MPa)	410	[46]
Coefficient of thermal expansion $\alpha$ ( $K^{-1}$ )		
	$55 \times 10^{-6}$	[58]
Density		
$\rho$ ( $kg/mm^3$ )	$1.3 \times 10^{-9}$	[8]

693

**Table 3**

Fibre–matrix interface properties

Initial stiffness	Interface strengths [60]		Interface fracture energy [20]		B-K law parameter [8]
$K$ (MPa/mm)	$t_1^0$ (MPa)	$t_2^0, t_3^0$ (MPa)	$G_{ic}^{int}$ ( $J/mm^2$ )	$G_{ilc}^{int}, G_{illc}^{int}$ ( $J/mm^2$ )	$\eta$
$10^8$	80	53	4	8	1.45

694

**Table 4**

Homogenized composite properties

$E_L$ (MPa)	$E_T$ (MPa)	$\nu_L$	$G_L$ (MPa)	$G_T$ (MPa)	$\alpha_L$ ( $K^{-1}$ )	$\alpha_T$ ( $K^{-1}$ )	$\rho$ ( $kg/mm^3$ )
168800	8796	0.3	4078	3180	$-0.12 \times 10^{-6}$	$37.5 \times 10^{-6}$	$1.59 \times 10^{-9}$

695

1 **Supplementary Material for:**

2 **Through-thickness crack growth resistance in fibre composites and its role in preventing ply**  
3 **cracking in cross-ply laminates**

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12  
13 **S1. Constitutive modelling of epoxy phase**

14 The epoxy matrix is modelled using an isotropic elastic-plastic with damage constitutive  
15 model proposed by Melro et al. [1]. In the present work, the epoxy model described in [1-5] is  
16 implemented as a VUMAT user subroutine in Abaqus [6]. The initial elastic behaviour is defined by  
17 a linear relation between the stress tensor and the elastic strain. Then, to appropriately capture the  
18 inelastic behaviour of epoxy polymers [7, 8] and the hydrostatic pressure dependency [8], this  
19 constitutive model is based on the paraboloidal yield and failure criteria, and uses a  
20 thermodynamically consistent damage model to predict damage growth. The paraboloidal yield  
21 criterion is defined as [1, 9]:

22 
$$f(\sigma, \sigma_c, \sigma_t) = 6J_2 + 2I_1(\sigma_c - \sigma_t) - 2\sigma_c\sigma_t, \quad (S1)$$

23 where  $\sigma$  is Cauchy stress tensor,  $J_2$  is the second invariant of deviatoric stress tensor,  $I_1$  is the first  
24 invariant of the stress tensor and  $\sigma_c$  and  $\sigma_t$  denote the tensile and compressive yielding stresses,  
25 respectively. A non-associative flow rule [1] is also introduced to correct the volumetric deformation

26 in plastic regime. The tensile and compressive hardening laws are defined as two piecewise functions  
 27 of the equivalent plastic strain. The hardening data of both tension and compression yield curves has  
 28 been extracted from the experimental results in [8], which characterizes the stress-strain behaviour of  
 29 a very typical Bisphenol-A type plain epoxy resin (i.e., Toho # 113) [8]. The damage onset of the  
 30 epoxy matrix is defined by a damage activation function [1]:

$$31 \quad F_m^d = \Phi_m^d - r_m = 0, \quad (S2)$$

32 where  $r_m$  is an internal variable related with the damage variable  $d_m$ , while the damage activation  
 33 variable,  $\Phi_m^d$ , is defined as [1]:

$$34 \quad \Phi_m^d = \frac{3\tilde{J}_2 + \tilde{I}_1(X_m^c - X_m^t)}{X_m^c X_m^t}. \quad (S3)$$

35 The invariants  $\tilde{J}_2$  and  $\tilde{I}_1$  are determined using the effective stress tensor as defined in [1], while  $X_m^c$   
 36 and  $X_m^t$  denote the ultimate compressive and tensile strengths. The internal variable  $r_m$  is defined as  
 37 [1]:

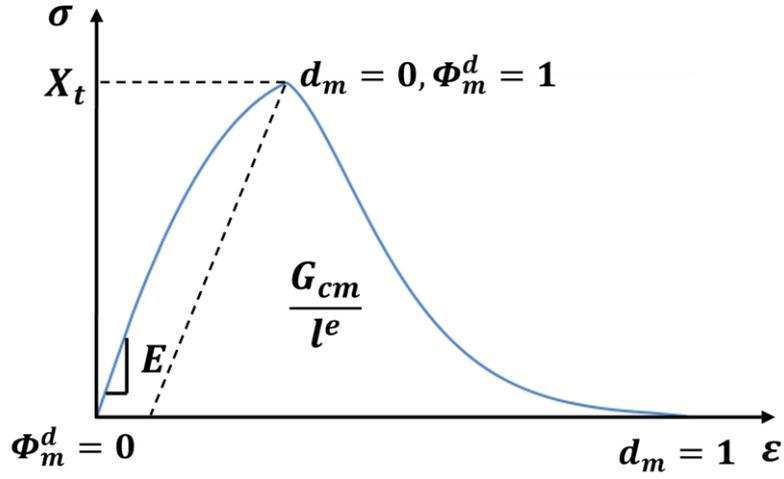
$$38 \quad r_m = \max \left\{ 1, \max_{t \rightarrow \infty} \{ \Phi_{m,t}^d \} \right\}. \quad (S4)$$

39 The relationship between  $r_m$  and  $d_m$  is given by an exponential damage evolution law [1, 5] which  
 40 is implemented along with the Bažant's crack band model [10] to mitigate the mesh size dependency  
 41 due to the material softening:

$$42 \quad d_m = 1 - \frac{e^{A_m(3 - \sqrt{7+2r_m^2})}}{\sqrt{7+2r_m^2} - 2}, \quad (S5)$$

43 where  $A_m$  is the parameter determined by conducting the regularization of the computed energy  
 44 dissipation of elements [1]. This involves using the fracture energy of the epoxy matrix,  $G_{mc}$  and the  
 45 characteristic element size,  $l^e$ , according to the Bažant's crack band model [10]. From the above  
 46 definition of the damage model (i.e., Eqs. (S3-5)), it is noteworthy that (i) when the epoxy matrix is  
 47 in an undamaged condition,  $0 \leq \Phi_m^d < 1$  and  $r_m = 1$ , leading to  $d_m = 0$ ; (ii) when the damage

48 criterion has been activated,  $\Phi_m^d \geq 1$  and  $r_m = \Phi_m^d$ , leading to  $0 \leq d_m \leq 1$ ; (iii)  $d_m = 1$   
 49 corresponds to when the material is fully damaged. More details about the computational  
 50 implementation of the constitutive model are presented in [1, 5]. The stress-strain response of the  
 51 epoxy model is schematically illustrated in Fig. S1 and the relevant material properties are listed in  
 52 Table 2 of the main paper.



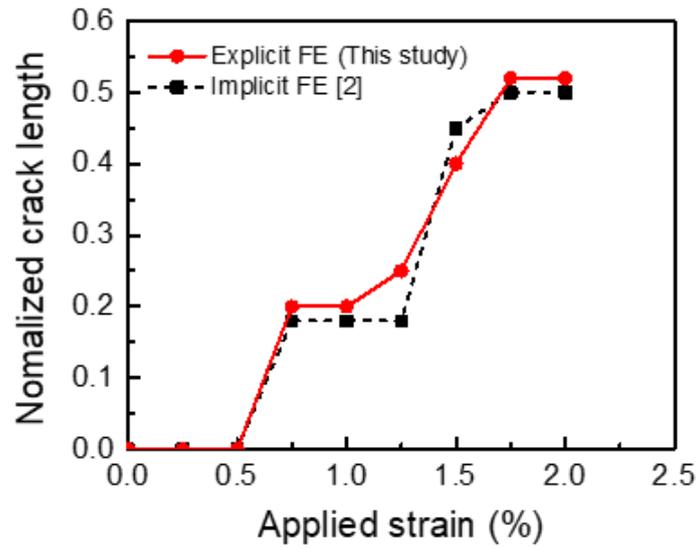
53

54 **Fig. S1** Schematic of the stress-strain response of the epoxy matrix.

55

56 **S2. Verification of the epoxy model**

57



58

59 **Fig. S2** Comparison between the present explicit approach and the implicit implementation in [4],  
 60 with respect to the normalized crack length in an inner 90° layer of thickness 20 μm and outer layers  
 61 of thickness 75 μm.

62

63 **S3. Interlaminar properties**

<b>Table S1</b>					
Interlaminar properties for the interfaces between inner and outer layers [4]					
Initial stiffness	Interface strengths		Interface fracture energy		B-K law parameter
$K$ (MPa/mm)	$t_0^n$ (MPa)	$t_0^s$ (MPa)	$G_{int}^n$ (J/mm <sup>2</sup> )	$G_{int}^s$ (J/mm <sup>2</sup> )	$\eta$
10 <sup>8</sup>	93	71	0.277	0.788	1.634

64

65

66 **S4. Estimation of the crack-growth resistance from the crack opening profiles**

67 The crack-growth resistance  $G^R$ , based on the tip-to-tip crack length  $2a$  of the **through-**  
68 **thickness matrix crack** and the maximum COD  $\delta_{max}$  for various levels of strain, as indicated by the  
69 crack opening profiles in Fig. 2(c) of the main paper, is calculated by the following formula:

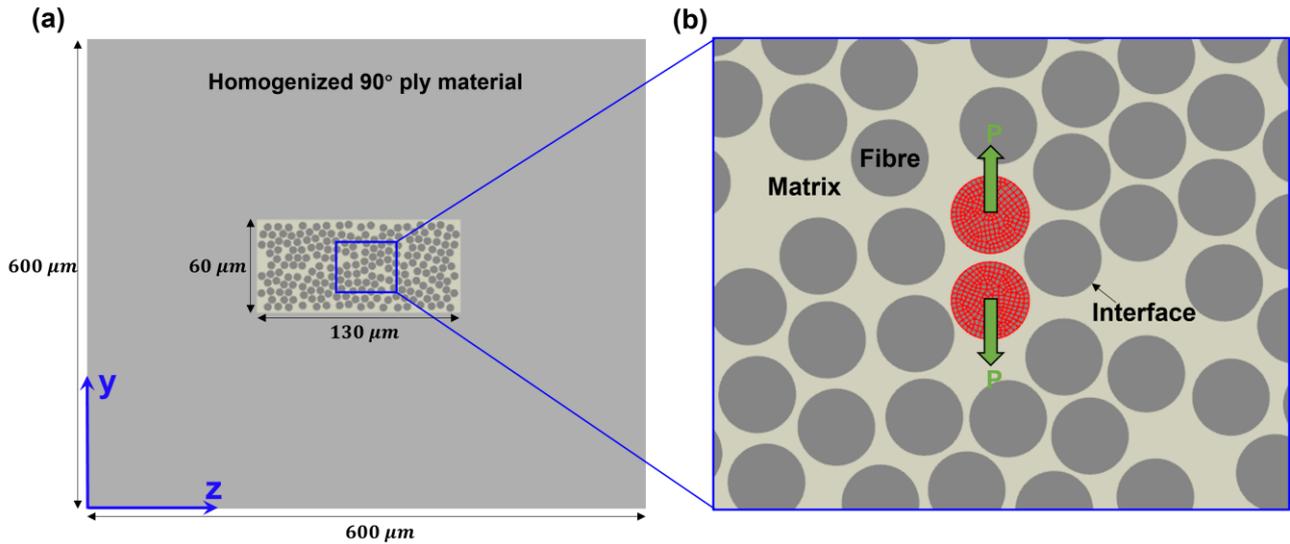
70 
$$G^R = \frac{\pi E_T}{16(1 - \nu_T^2)} A^2 a,$$

71 
$$A = \frac{\delta_{max}}{a},$$
 (S6)

72 where  $E_T = 8796 \text{ MPa}$  and  $\nu_T = 0.32$ , denoting the transverse elastic modulus and Poisson's ratio  
73 of the uncracked lamina in the present study.

74

75 **S5. Initial attempt on generating the R-curve using two neighbouring loading fibres**



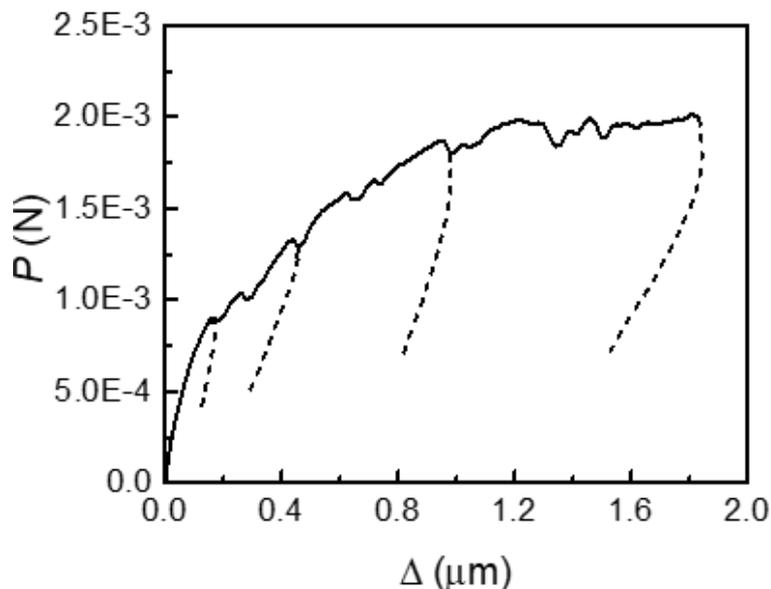
77 **Fig. S3** Configuration of the model for initial attempt on generating the R-curve using two  
78 neighbouring loading fibres.

79 As mentioned in Section 3 of the main paper, the finite element (FE) model for the initial  
80 attempt on generating the R-curve computationally is shown by Fig. S3(a). The entire model consists  
81 of 90° ply material only. For computational efficiency, a rectangular micromechanical model is

82 embedded within a square region. The same material properties, mesh size and element types as  
83 described in Section 3 of the main paper are employed for both the homogenized (elastic) region and  
84 the micromechanical model region. The displacement field is assumed to be continuous across the  
85 interfaces between the embedded cell and the homogenized outer portions. The model thickness in  
86 the  $x$ -direction (the fibre axis) is twice the average element size within the embedded cell, and periodic  
87 boundary conditions are applied to the faces normal to the  $x$ -axis.

88 There is no pre-existing crack in this model. To initiate the matrix cracking in a natural manner  
89 at the fibre-matrix interface and thus to generate the R-curve with the progression of the matrix crack,  
90 two neighbouring fibres, with a relatively small inter-fibre spacing, which are aligned with the  $y$ -axis  
91 direction are chosen to be the load application points as shown in Fig. S3(b). All nodes of the upper  
92 loading fibre are given a specified vertical displacement  $u_y$ , whereas all nodes of the lower loading  
93 fibres are held fixed. The resulting nodal forces for the upper loading fibres are summed to obtain the  
94 force  $P$ , whereas the displacement is given by  $\Delta = u_y$ .

95

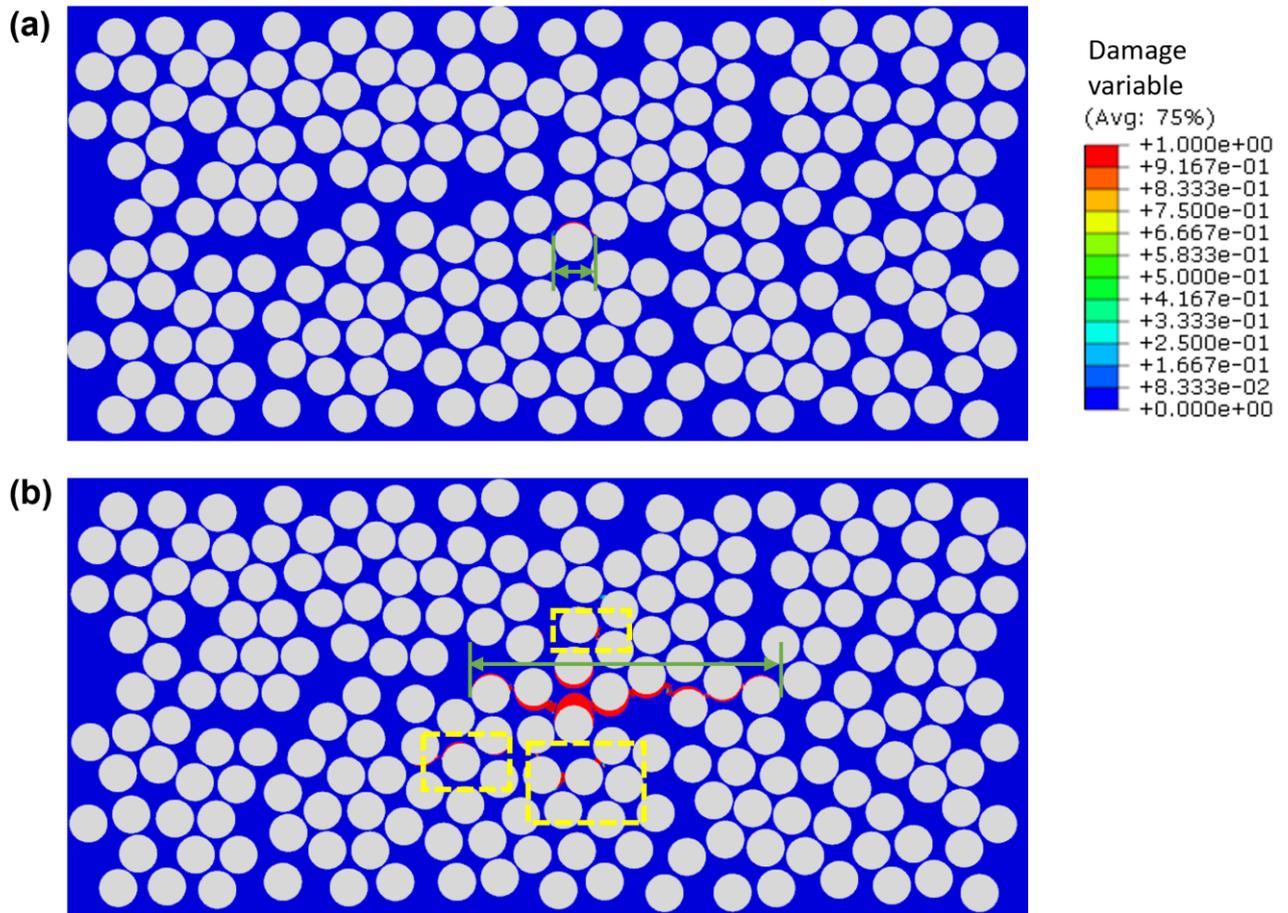


96

97 **Fig. S4** The load-displacement plot.

98

99           The load-displacement ( $P - \Delta$ ) curve obtained in this manner is shown in Fig. S4. This curve  
100 includes some periodical unloading curves that do not extrapolate back to the origin, indicating that  
101 model response is not linearly elastic. The contour plot of damage variable in Fig. S5(a) shows that  
102 the microcracking does indeed initiate at the fibre-matrix interface (i.e. debonding) with the tip-to-tip  
103 length of the matrix crack  $2a$  indicated by the green arrow, which is in accord with the previous  
104 experimental and computational observations [4, 11, 12] as well as the present micromechanical  
105 simulation results in Fig. 2(b) of the main paper. With continuing to apply the load, however, some  
106 matrix damages appear in the direction of loading, as indicated by the yellow dashed box in Fig S5(b),  
107 which are not directly associated with the main crack growth process that is transverse to the loading  
108 direction. These damages are mainly caused by the compressive and shear deformation of the matrix  
109 near the fibres, due to the squashing effect caused by the upper and lower loading fibres, which does  
110 not conform to the intended application of the R-curve for the context of interest. Furthermore, Fig.  
111 S6 shows the plastic zone generated by this approach of loading. It is now clear that the severe non-  
112 associated plastic deformation and damage caused by the loading fibres along the direction of loading  
113 are responsible for the inelastic unloading response shown in Fig. S4, and the contribution of this  
114 extraneous energy dissipation to the work of fracture could not readily be quantified.

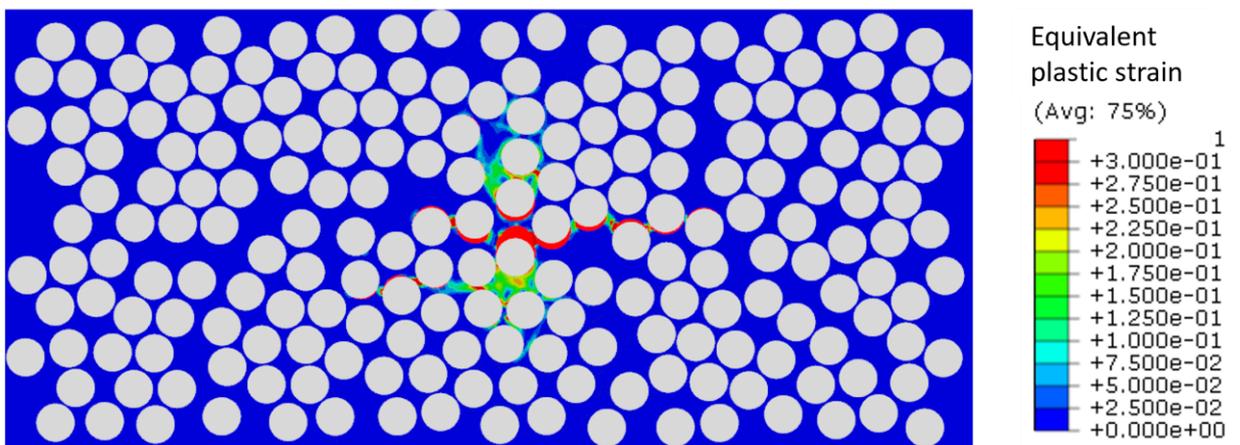


115

116 **Fig. S5** Contour maps of the damage variable at the tip-to-tip length of the matrix crack of (a)  $5 \mu m$

117 and of (b)  $40 \mu m$ .

118



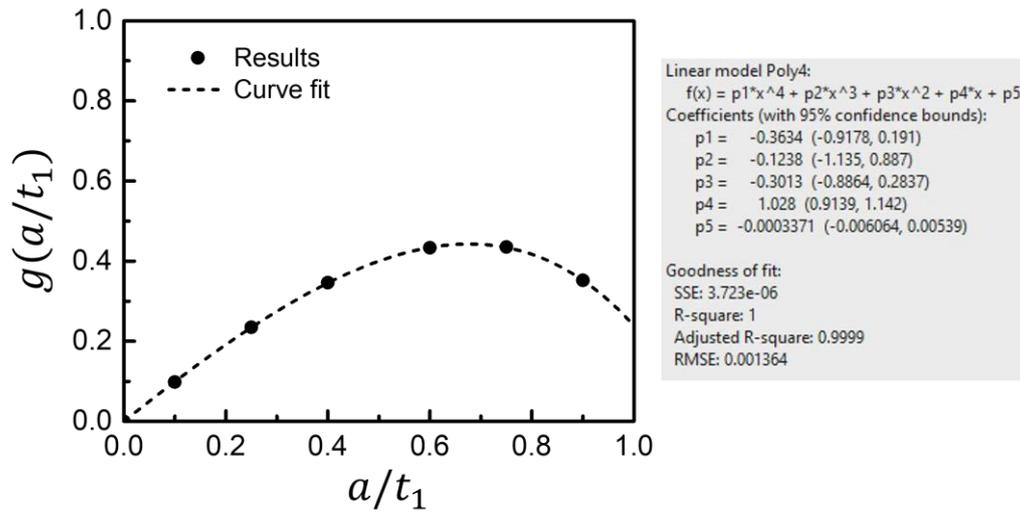
119

120 **Fig. S6** Contour maps of the equivalent plastic strain at the tip-to-tip length of the matrix crack of

121  $40 \mu m$ .

122 **S6. Curve fit of the results for  $g(a/t_1)$ , for  $E_T/E_L = 0.05$**

123

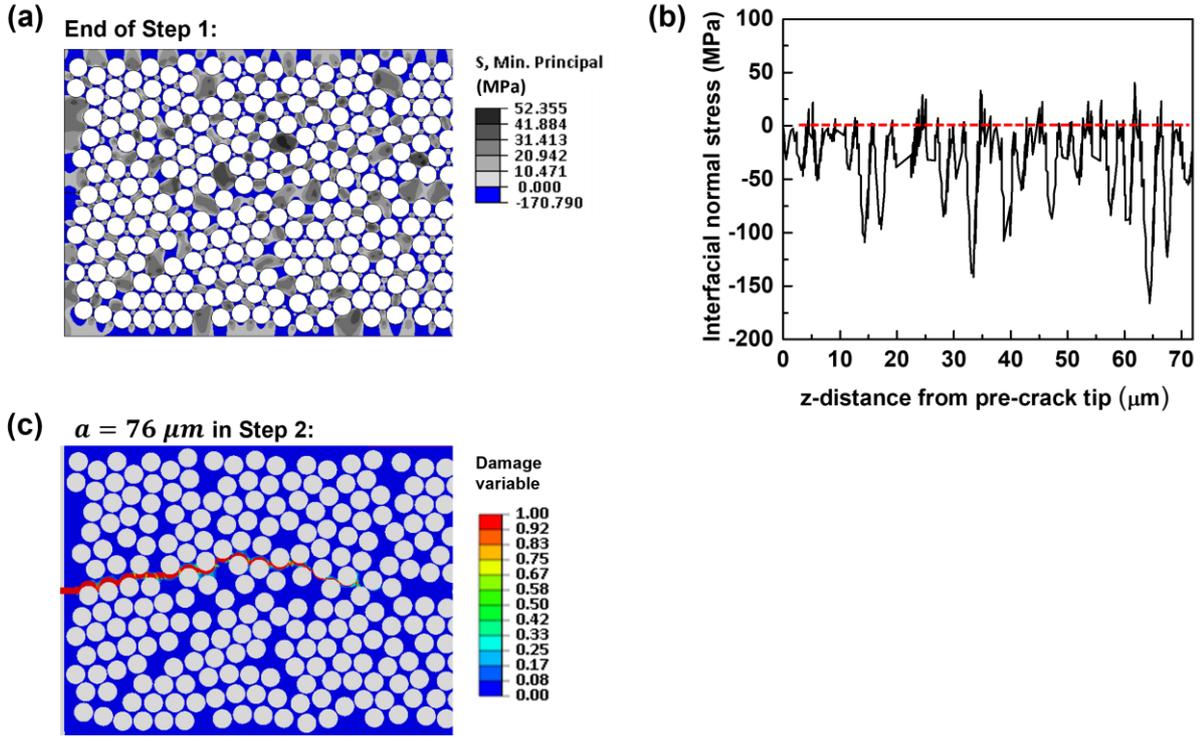


124

125 **Fig. S7** Fourth-order polynomial curve fit of the results for  $g(a/t_1)$  in  $a/t_1$ , for  $E_T/E_L = 0.05$ , along  
126 with the curve-fit quality analysis done by using MATLAB [13].

127

### S7. Intra-ply thermal residual stresses at the completion of Step 1



129

130 **Fig. S8** (a) The intra-ply thermal residual stresses field at the completion of Step 1. (b) The interfacial  
 131 normal stress at the completion of Step 1, along the prospective crack path as (c) the crack of length  
 132 of 76 μm generated in Step 2.

133

### 134 S8. Laminate analysis for calculating the thermal residual strain at the ply level, $\varepsilon_{yy}^{th}$

135 Consider the cross-ply laminate shown in Fig. 6(a) of the main paper, which has undergone a  
 136 quenching process (Step 1) with a resulting temperature change  $\Delta T = -278K$  as given in Section  
 137 5.1 of the main paper, the thermal strains in a ply are [14]:

$$138 \{\varepsilon\}_{12}^T = [\theta]^{-T} \{\alpha_L\} \Delta T - \{\alpha_P\} \Delta T, \quad (S7)$$

139 where the subscripts "12" denote quantities in the local material coordinate (i.e. fibre direction=1,  
 140 lateral direction=2),  $\alpha$  the coefficient of thermal expansion (CTE). The subscripts "P" and "L" signify  
 141 quantities pertinent to the ply and laminate, and  $[\theta]^{-T}$  the transfer matrix given by:

142

$$[\theta]^{-T} = \begin{bmatrix} m^2 & n^2 & mn \\ n^2 & m^2 & -mn \\ -2mn & 2mn & m^2 - n^2 \end{bmatrix},$$

$$m = \cos(\theta), n = \sin(\theta). \quad (S8)$$

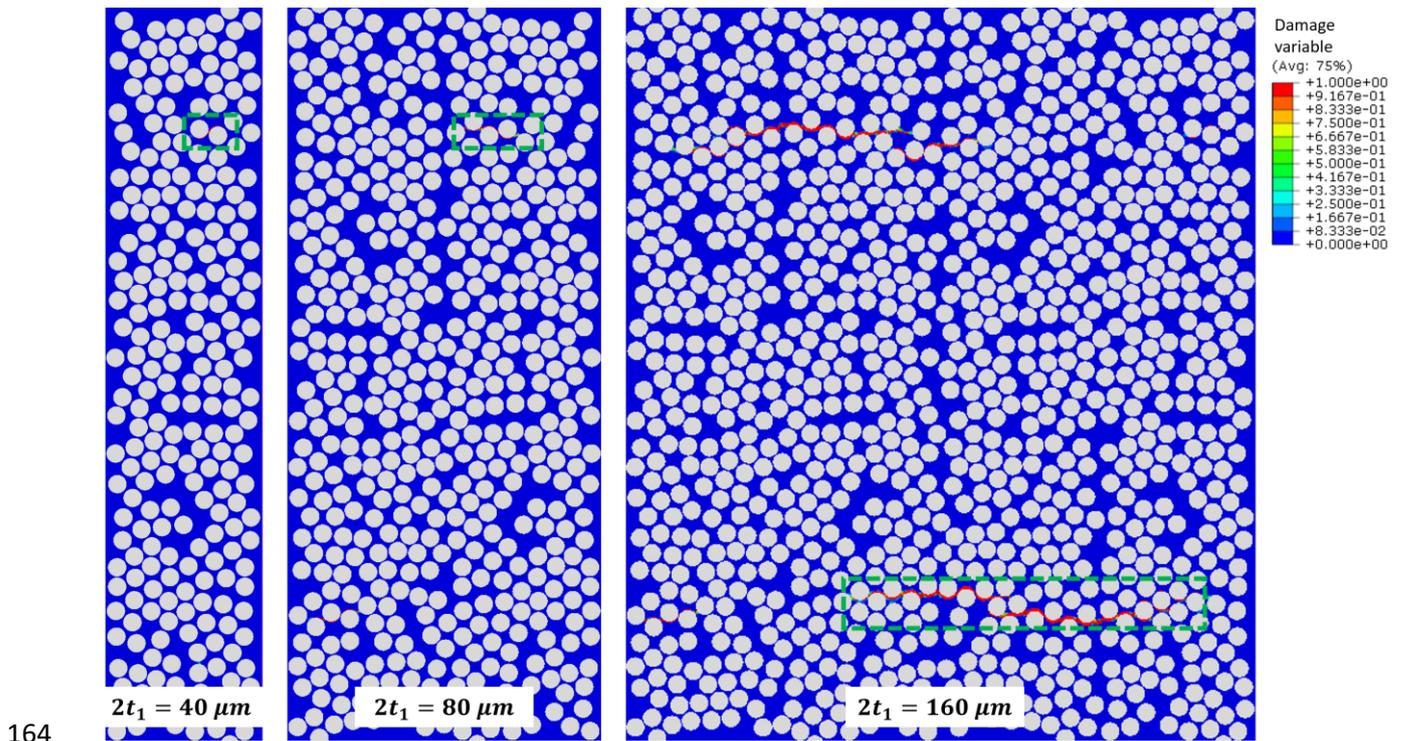
For the inner  $90^\circ$  ply,  $\theta = 90^\circ$ . Having known the material properties of a ply including CTEs  $\{\alpha_L\}$  (see Table 4 of the main paper), the laminate CTEs  $\{\alpha_P\}$  can be calculated by using conventional lamination theory [15]. Finally, the lateral thermal residual strain  $\varepsilon_{yy}^{th}$  incorporated in Eq. (4) of the main paper is:

$$\varepsilon_{yy}^{th} = \varepsilon_{22}^T, \quad (S9)$$

where  $\varepsilon_{22}^T$  is the component of  $\{\varepsilon\}_{12}^T$  pertinent to the lateral strain.

In Figs. 9(a-c) and Figs. 10(a-c) of the main paper, the dotted vertical lines represent the thermal strain on  $90^\circ$  layer  $\varepsilon_{yy}^{th}$  at the completion of Step 1. There is little change in the value of  $\varepsilon_{yy}^{th}$  for various inner layer thickness:  $\varepsilon_{th} = 1.04\%$  for  $2t_1 = 40 \mu m$ , and  $\varepsilon_{th} = 1.03\%$  for  $2t_1 = 160 \mu m$ . This lack of sensitivity to the inner layer thickness is partly the consequence due to the relatively thick outer layer thickness employed in the present laminate configuration to conform to the experimental setup by Saito et al. [11]. Nevertheless, these values of inter-ply residual strain exceed the failure-initiation strain for fibre-matrix debonding, particularly when  $2t_1 = 160 \mu m$  for which even the full-thickness cracking is predicted to occur when  $G_c = 60 J/m^2$ , i.e., no applied strain is required to cause ply cracking, as can be seen from Fig 9(d) of the main paper. The representative contours illustrating the extent of matrix cracking for baseline matrix toughness  $G_c = 120 J/m^2$  at the completion of Step 1 of the micromechanical simulations are presented below in Fig. S9.

163



164

165 **Fig. S9** Contour maps of the damage variable at the completion of Step 1 for various inner 90° layer  
 166 thicknesses:  $2t_1 = 40, 80$  and  $160 \mu m$  (the through-thickness cracks that are selected for measuring  
 167 the crack length are indicated by the green dashed rectangles, and only the inner 90° layer is shown).

168

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