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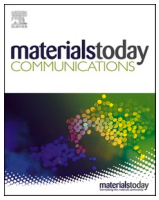

Vacuum-infused thermoplastic fibre-metal laminates – Advances in bonding and recycling

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ABSTRACT

Novel methods for bonding and recycling acrylic fibre-metal laminates are investigated. Interfacial bonding is achieved via a methacrylate-based adhesive film. Short beam shear testing was performed to assess the bond quality in like-for-like comparisons of different test parameters. It is shown that apparent interlaminar shear strength is influenced by the adhesive carrier and the initiator. The latter is corroborated by micrographs showing a transition from adhesive failure to cohesive failure. It is confirmed that the adhesive is dissolved by liquid acrylic resin during infusion. Dissolution is used to debond the fibre-metal interface for recycling.

1. Introduction

Fibre-metal laminates (FMLs) combine layers of composite and metal to achieve a unique set of material properties that balance the advantages and disadvantages of the constituent materials [1]. Originally developed for the aerospace industry, FML technology has the potential to expand into other industries, such as wind energy and marine transport, when used in combination with new reactive thermoplastic systems like Elium® [2–4]. Elium®, is a low viscosity resin that can be polymerised at ambient temperatures to form a polymethylmethacrylate (PMMA a.k.a. acrylic) matrix. One critical aspect of combining these technologies is optimising the metal-composite interface [1,5,6]. As the constituent materials are dissimilar, weak interfacial bonding may lead to delamination and a loss of structural integrity [7]. Interlaminar shear strength (ILSS) is an important metric for mechanically evaluating the interfacial bond. Due to its simplicity and efficiency, the short beam shear (SBS) test method is the most widely used technique for measuring the apparent ILSS of FMLs [7]. Despite several caveats to the technique [8,9], the SBS method is useful for assessing bond quality in like-for-like comparisons e.g. comparison between specimens with different surface treatments [10] or adhesive types [11]. To date, few studies have investigated methods for improving interfacial bonding between Elium matrix composites and metals [10–12]. Initial studies focused on the use of surface treatments to optimise the metal surface for bonding [10,12], while more recent studies have investigated the use of various adhesive interlayers between the metal and composite [11]. It has already been shown that the solubility of this thermoplastic matrix allows for greater recyclability [13,14], but, to the best of the authors' knowledge, there has been no research published on recycling of FMLs via dissolution. In this study, several glass veils are evaluated as adhesive carriers, the adhesive initiator is investigated in terms of its effect on bonding strength, and the dissolution of the adhesive layer is investigated both in terms of bonding and debonding.

2. Experimental

2.1. Materials

Elium® 188 O resin (ARKEMA, France) was used with BP-50-FT initiator (United Initiators) in a 100:3 wt ratio. The fibre reinforcement was a 646 g/m^2 stitched, unidirectional (UD) glass fibre (GF) fabric (Ahlstrom-Munksjö), which had a multicompatible sizing. The metal layer was a 6082-T6 grade aluminium (0.71 mm thick), anodised in sulphuric acid (NPI Solutions). A methacrylate adhesive, SAF 30–5 (Bostik), was used to improve bonding at the fibre-metal interface. This adhesive was supplied in two parts: resin and initiator. A nonwoven GF carrier (Technical Fibre Products Ltd.) was combined with the adhesive to control the bondline thickness and maintain its uniformity [15].

Initially, three GF carriers with different areal weights were trialled; 6, 17, and 34 g/m^2 (see Table 1). For these trials, the adhesive resin was applied without an initiator, as described by Robert et al. [11]. The assumption was that, as the FML was infused with Elium® 188 O and BP-50-FT, the latter would initiate polymerisation in the adhesive. Following this, one GF carrier was downselected for further trials to

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Table 1
Material test parameters. The test IDs denote the combination of test parameters.

Test ID	Areal weight [g/m^2]	Initiator type	Initiator content [%]
34-A-0	34	-	0.0
17-A-0	17	-	0.0
6-A-0	6	-	0.0
6-B-1.5	6	BP-50-FT	1.5[1]
6-B-3	6	BP-50-FT	3.0[1]
6-C-10	6	SAF 30–5 (initiator)	10.0[2]

[1] weight percentage
[2] volume percentage

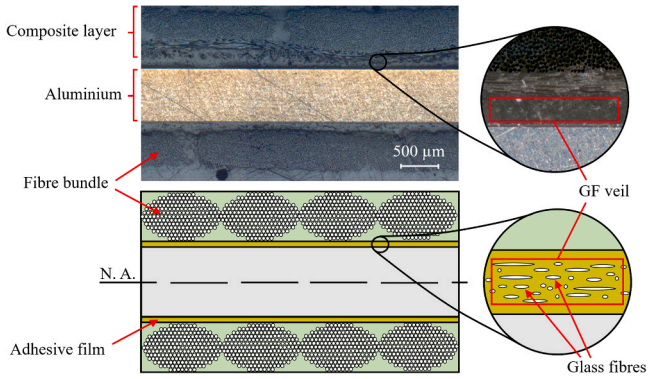

Fig. 1. (Top) Optical micrograph of the FML cross-section. (Bottom) Schematic of the FML indicating the location of the adhesive layer with its embedded GF veil.

investigate the effect of dosing the adhesive resin with initiator prior to infusion. Two types of initiator were investigated, BP-50-FT and the initiator supplied with SAF 30–5; each mixed in a specific weight/volume ratio, also shown in Table 1. FMLs were manufactured using a standard vacuum infusion process (VIP). The stacking sequence of each FML was [0/Al/0], as shown in Fig. 1.

The mean fibre volume fraction (FVF) of the composite layer was estimated to be 41.95% ± 1.63%. This was determined using the matrix burn-off method (Procedure G – ASTM D3171–15 [16]). The glass transition temperature (T_g) was measured as an additional control check using dynamic mechanical thermal analysis (DMTA – Triton 2000). In single cantilever mode, with an oscillating displacement of 0.03 mm at 1 Hz and a ramp rate of 3 °C/min, the mean T_g was measured as 104.96 °C ± 3.48 °C (tan delta peak).

2.2. Methods

Short beam shear (SBS) testing was carried out in accordance with BS ISO 14130:1998. Tests were performed using an Instron 3369 test machine with a 10 kN load cell, with a span-to-thickness ratio of 5 – specimens had a mean thickness of 2.6 mm.

Scanning electron microscopy (SEM) was performed on the metal-composite interface of failed SBS specimens (note, the specimens were not sputter-coated). Micrographs were captured using a Hitachi TM4000Plus Tabletop Microscope. The backscattered electron (BSE) detector was used to contrast the residual adhesive left on the aluminium surface, while the secondary electron (SE) detector was used to capture details of the adhesive film surface. Accelerating voltages of 15 kV and 5 kV, respectively, were used.

Two tests were performed to investigate the dissolution of the polymerised adhesive layer. The purpose of these tests was to:

- Determine whether or not the adhesive was dissolved during infusion of the FML
- Determine if the FML could be debonded for recycling

For test 1, the adhesive resin was mixed with the supplied SAF 30–5 initiator and applied to several aluminium adherends. After 24 h, the mass of each sample was measured using an analytical balance and then immersed in Elium® 188 O resin for 1–20 min before being removed and weighed again.

For test 2, an FML sample (6-C-10) was immersed in Elium® 188 O resin and stirred at several intervals over a 24 h period. After 24 h, the sample was removed and tweezers were used to separate the composite layer from the aluminium.

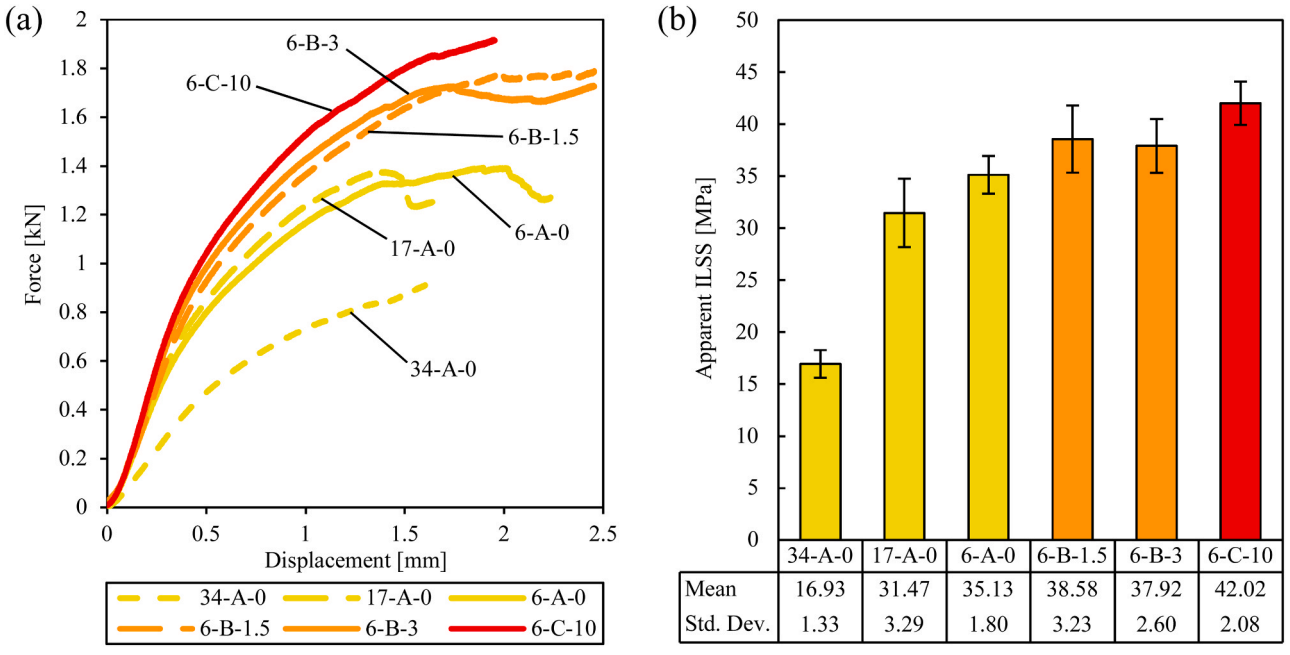

Fig. 2. Results of each SBS test case. (a) Representative force-displacement data. (b) The mean apparent ILSS, with error bars representing ± 1 standard deviation.

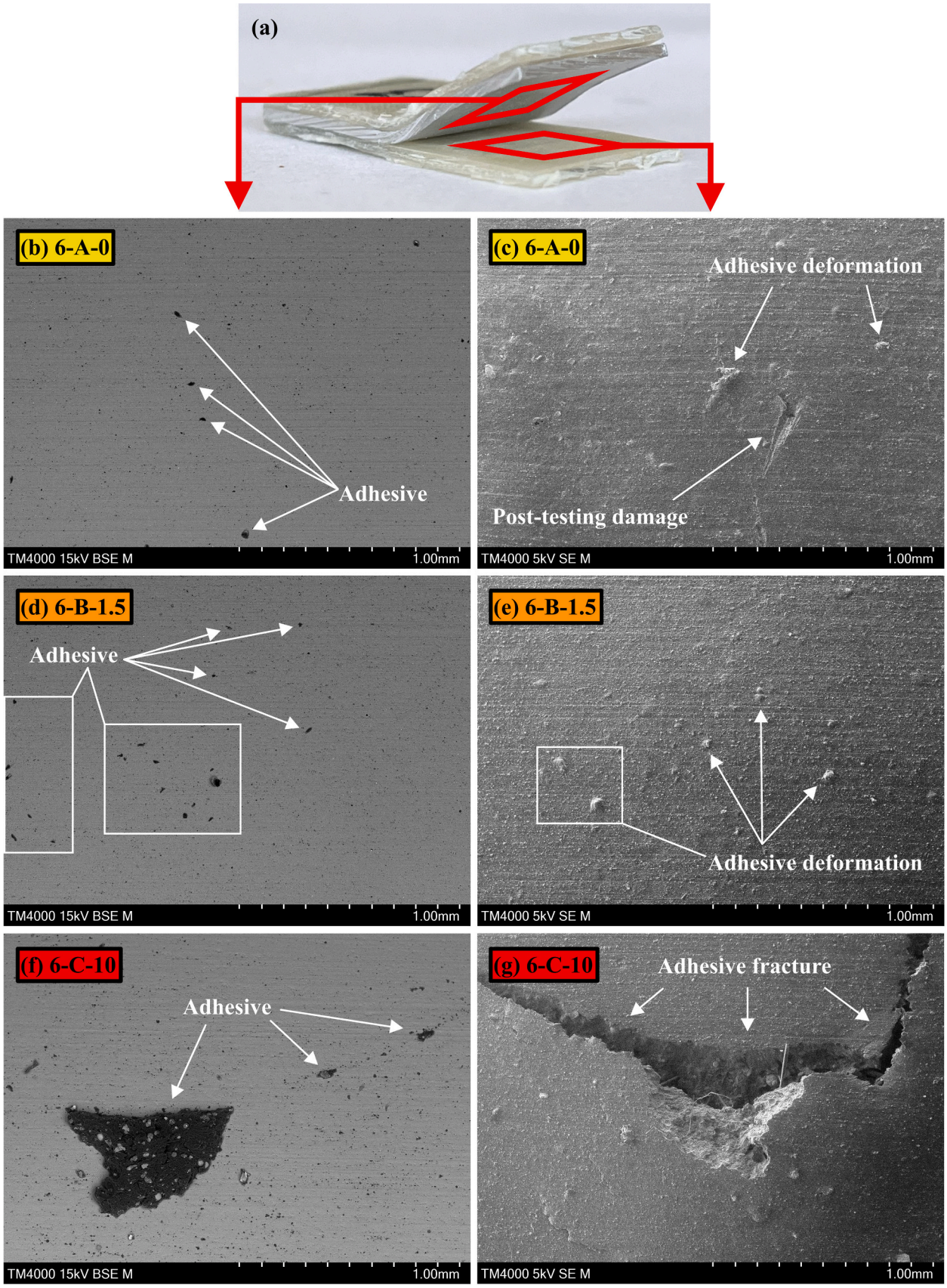

Fig. 3. (a) Failed SBS specimen indicating the location at which SEM micrographs were captured. (b, d, and f) Residual adhesive bonded to the aluminium surface, scanned using backscattered electrons (BSE) at 15 kV. (c, e, and g) Secondary electron (SE) scanning of the adhesive layer (at 5 kV) showing localised deformation (c and e) and fracture (g). All micrographs are × 150 magnification.

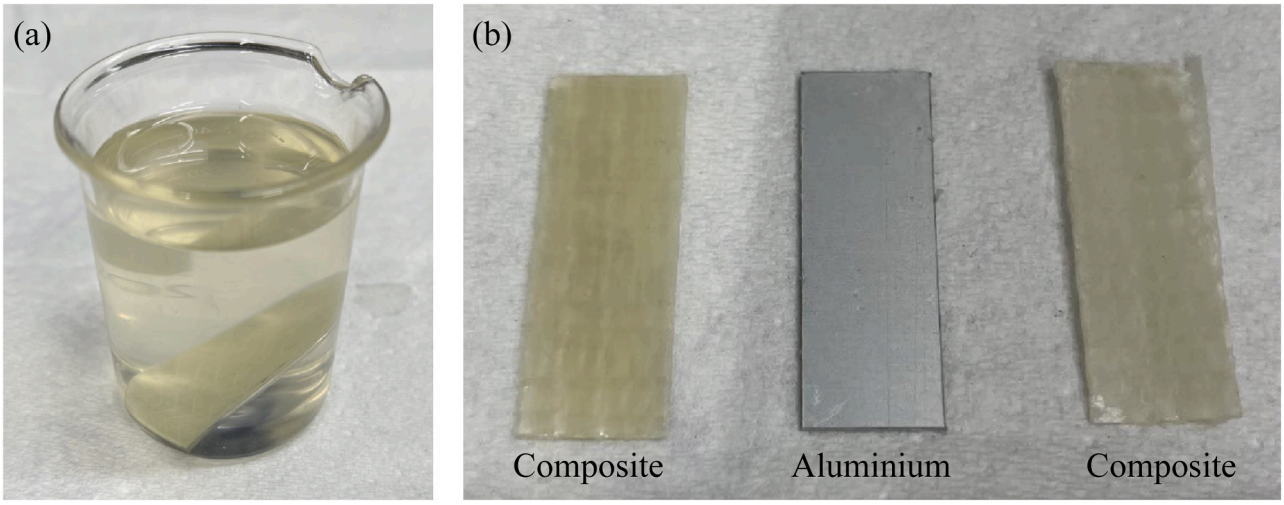

Fig. 4. Results of the debonding trial: (a) FML sample (6-C-10) placed in Elium 188 O resin; (b) The aluminium fully debonded from the composite after 24 h.

3. Results and discussion

3.1. Apparent interlaminar shear strengths

Fig. 2 shows representative force-displacement data for the FML SBS specimens alongside and their apparent interlaminar shear strength (ILSS). Focusing on the results of the GF carrier trial first (i.e. 34-A-0, 17-A-0, and 6-A-0), the ILSS was found to increase as the areal weight of the GF carrier decreased. This trend was expected as tough, ductile adhesives perform better in shear when forming a thin, uniform bondline [15,17]. The reliable formation of uniform bondlines will be important for translating this technology to an industrial scale. Poor bondline formation results in a significant drop-off in ILSS e.g. 34-A-0 specimens. This drop-off corresponded with an increased presence of voids in the adhesive layer; see Figure A.1 in the Supplementary material. These voids formed due to poor impregnation of the GF carrier (note, SAF 30–5 has a high viscosity and short open time; approximately 140 Pa s and 7 min, respectively). In contrast, the ILSS of the 6-A-0 specimens was approximately the same as reported by Robert et al. for the same material system (i.e. 35.4 MPa [11]). This indicated that the 6 g/m^2 GF carrier did not have any adverse effect on bonding.

In their previous work, Robert et al. [11] assumed that the initiator content of the Elium® resin (3 wt%) was sufficient to polymerise the adhesive layer after infusion, however, they did not test the concept of dosing the adhesive film with initiator prior to infusion. Given that acrylics are amorphous in nature and highly susceptible to dissolution, acrylic monomers can dissolve acrylic polymer and then be polymerised to form a new polymer network [14]. Recently, it has been demonstrated by Roy et al. [18] that this phenomenon can be exploited to join acrylic polymer composites. They found that the molecular entanglement created by this process increased the ILSS by approximately 23%. In a similar manner, the study presented herein investigated dosing the adhesive layer with initiator prior to infusion with Elium®. As shown in Table 1, two different types of initiator, BP-50-FT and SAF 30–5, part B, were investigated, with two different weight percentages used for the former; 1.5 wt% and 3 wt%. Note, BP-50-FT was the initiator used to polymerise Elium® 188 O, while SAF 30–5, part B was the initiator supplied with the SAF 30–5 adhesive resin (part A). Fig. 2 shows that, similar to the findings of Roy et al. [18], dosing the adhesive layer with initiator prior to infusion resulted in a corresponding increase in ILSS; up to 20% from the ILSS of 6-A-0. Moreover, the type of initiator had an effect on the bond strength, whereas the initiator content did not (i.e. the ILSS for 1.5 wt% and 3 wt% of BP-50-FT were approximately the same). Acceptable interlaminar shear failure was observed for each test specimen, however, in the case of 6-C-10, the failure transitioned from end-opening to local shear debonding i.e. from full delamination to partial delamination. This suggested that dosing with the SAF 30–5 initiator provided better interfacial bonding with the aluminium than BP-50-FT.

3.2. Failure surface morphology

The change in ILSS corresponded with a visible change in the morphology of the failure surface. For closer inspection, SEM was performed on the failure surfaces of the 6-A-0, 6-B-1.5, and 6-C-10 specimens. Fig. 3 shows representative SEM micrographs for each.

As can be seen in Fig. 3(b, d, and f), the increasing ILSS corresponded with increasing quantities of residual adhesive on the aluminium surface. For the 6-C-10 specimens (Fig. 3(f and g)), fractures in the adhesive layer signalled a transition to cohesive failure. In contrast, Fig. 3(c and e) showed only localised deformation of the adhesive for specimens 6-A-0 and 6-B-1.5. Most likely, this was a consequence of the pitted aluminium oxide surface formed by the anodisation process [10]. This pitting allowed for localised mechanical interlocking of the adhesive layer and the aluminium oxide surface.

3.3. Adhesive layer dissolution

To investigate dissolution of the adhesive layer, two trials were performed as described in Section 2.2. Despite having been fully polymerised, the first trial found that dissolution of the adhesive was occurring in less than 1 min. This confirmed that the adhesive was dissolved during infusion, thus allowing monomer and initiator to diffuse through the adhesive polymer and polymerise to form a new polymer network.

The second trial was performed to investigate dissolution for the purpose of debonding and recycling. As shown in Fig. 4, after immersion in Elium® 188 O, it was possible to separate the aluminium from the composite with little force required. Naturally, this has large implications for end-of-life recycling [13,14], as well as repairability and the potential for self-healing [19].

4. Conclusions

Advancements in the bonding and recycling of vacuum-infused thermoplastic fibre-metal laminates have been presented. It has been shown that a thin GF veil can be introduced as an adhesive carrier to improve bondline uniformity without compromising the interlaminar shear strength. The interlaminar shear strength was increased by dosing the adhesive with initiator prior to infusion of the composites layers. The

type of initiator used had an influence on the interlaminar shear strength, however, the initiator content did not. The increased interlaminar shear strength was linked to the dissolution of the adhesive during infusion prior to formation of a new polymer network with increased molecular entanglement. It has been shown that the acrylic matrix of the finished FML can also be dissolved to separate the aluminium and composite for recycling.

CRediT authorship contribution statement

James M. Maguire: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Visualisation, Project administration. **Vasileios Koutsos:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition**. Dipa Ray:** Conceptualization, Methodology, Resources, Writing – review & editing, Supervision, Project administration.

Declaration of Competing Interest

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

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.mtcomm.2023.106961.

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