Dose control of double-sided irradiation for polymer-matrix composites fabricated by in-situ curing with low energy E-Beam

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Abstract

The electron beam with energies lower than 150keV is applicable in in-situ curing of polymer-matrix composites but poor in penetration ability, leading to significantly attenuation of irradiation dose along the prepreg thickness. Experimental results in this paper show that laminates fabricated by prepregs single-sided irradiated are low in ILSS owing to incomplete curing at 180 ℃ for 30min. A double-sided irradiation method which includes equal dose and different doses on both sides of the prepreg is proposed to conquer this difficulty. Analysis show that under a total dose of 115kGy, 130kGy, 145kGy and 160kGy, the ILSS results get a biggest increase of 10.6%, 15.9%, 17% and 37.7% respectively by tuning the dose differences between both sides of the prepregs. Larger dose difference decreased the ILSS of the laminate because of the weakened physical and chemical bonding owing to higher curing degree and larger curing synchronicity difference.

1. Introduction

With the wide applications of resin matrix composites in the aerospace, energy and transportation industries[1,2], various curing process have been put forward to overcome the disadvantages of traditional thermal curing process[3-10]. Among them, the E-Beam curing technology has drawn much attention owing to its high curing efficiency, low energy consumption and high controllability[11-14]. Generally, the electron beam used for curing the composite materials is divided into high energy electron beam (5-15MeV) and low energy electron beam (below 300 keV). There has been much research on the high energy electron beam curing process and aircraft manufacturers have some successful cases of applications to fabricate or repair the composite components[15-17]. However, disadvantages of expensive and heavy shielding system and high irradiation limit the further development of the high energy electron beam curingtechnology. Compared with the high energy electron beam curing process, the low energy electron beam has lower risk of radiation, lower shielding cost and flexible operability. Cirri G[18] proposed the method that combine in-situ low energy electron beam curing with fiber placement. F. Guasti and E. Rosi developed composite fabricating laboratory equipment combined wet winding technology and low energy electron beam curing processing and conducted preliminary experimental study[19]. Several research institutions including NASA conducted feasibility study on manufacturing method of large scale resin matrix composites by automated tape placement with in-situ electron beam curing using the experimental prototype in NASA Marshall Space Flight Center[20].

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To further reduce the low energy E-Beam irradiation and shielding cost and improve the operability during in-suit curing, Dilmurat Abliz[21] has carried out fundamental research on E-Beam curing with energies lower than 150keV. In this paper, three irradiating methods for dose on prepregs were investigated. The effect of the irradiation dose distribution on the ILSS of laminates fabricated by in-situ curing with the low energy electron beam was analyzed.

2. Experimental

2.1. Materials

T700/EB99-1 carbon fiber prepreg tape was purchased from Institute of Aeronautical Materials, China. The resin was bisphenol A epoxy resin with 1.5% concentration of cationic initiator, which was proved suitable for the electron beam curing. By absorbing the electron beam energy, the cationic initiators decomposed protonic acid H^+[22,23](show in equation 1) which then reacted with the oxygen atom of the epoxy groups[22,23]. The T700 carbon fiber was purchased from Toary Industries, Inc.

$$\begin{gathered} Ar_2I^+MF_6^- \xrightarrow{e^-} [Ar_2I^+MF_6^-] \\ [Ar_2I^+MF_6^-] \rightarrow ArI\bullet^+ + Ar^\bullet + MF_6^- \\ ArI\bullet^+ + R-H \rightarrow ArI^+H + R\bullet \\ ArI^+H \rightarrow ArI + H^+MF_6^- \end{gathered} \tag{1}$$

2.2. Fabrication of composite laminates

The low energy E-Beam emitter used in this study was e250H produced by Advanced Electron Beam, Inc.. The device was shielded with thin stereotype.(Fig.1).

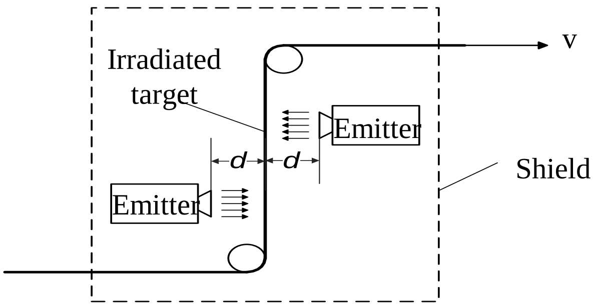

Figure 1. Low energy electron beam device

The fiber placement machine is developed by Xi'an Jiaotong University based on the FANUC robot. Fig.2 shows the fabricating process of the composite laminates.

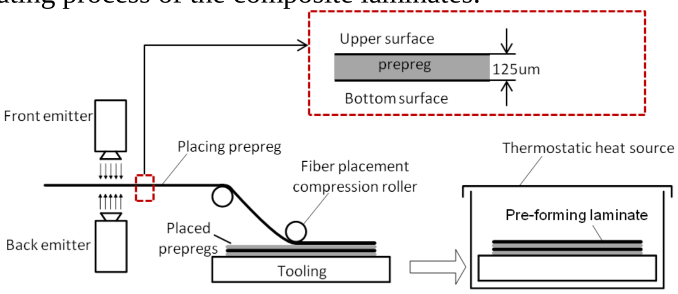

Figure 2. Composite laminates fabricating process

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2.3. Distribution of Dose

Fig.3 shows the irradiation dose distribution testing. According to Kanaya—Okayama[24] empirical formula, the penetration depth of electron beam in the prepreg tape and the dosimeter film has the relationship: $D_p/D_f=\rho_f/\rho_p$, where D_p and D_f are respectively the penetration depth of electron beam in the prepreg tape and dosimeter film, ρ_f is the density of the dosimeter film($1gcm^{-3}$)and ρ_p is the density of the prepreg tape($1.6g/cm^3$).

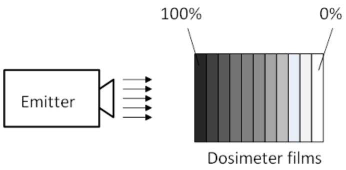

Figure 3. Irradiation dose distribution testing

2.4. Differential scanning calorimetry testing[25]

The curing degree of the prepreg tape was measured using differential scanning calorimetry (METTLER-TOLEDO).

2.5. ILSS test

The interlaminar shear strength (ILSS) of composite laminates was measured using an Instron 4467 universal testing machine and the three-point bending method was used according to ASTM D-2344 testing standard[26]. The specimens used for the short beam shear test were 18mm of length, 6mm of width and 3mm of thickness.

2.6. Microscopic features imaging

The cross-section of the experimental samples was observed using KEYENCE VH-600 optical microscope. The delamination surface of the experimental samples after ILSS test was observed using HITACHI S-3000 scanning electron microscope (SEM). The surface was coated with a thin evaporated layer of gold to improve conductivity before SEM observation.

3. Results and Discussion

3.1. Curing characteristics of prepregs

Fig.4 (a) shows the transmission characteristic of low energy E-Beam in the 125um thickness prepreg under 125keV. It's obvious that when the prepreg was irradiated from one side, dose decayed significantly along the prepreg thickness direction and remains almost no dose on the un-irradiated side of the prepreg, which means that the degree of cure along the thickness direction was also nonuniform. While the prepreg was irradiated with equal dose on both sides, accumulated dose turns out to be almost uniform along the prepreg thickness direction, shown in Fig.4 (b).

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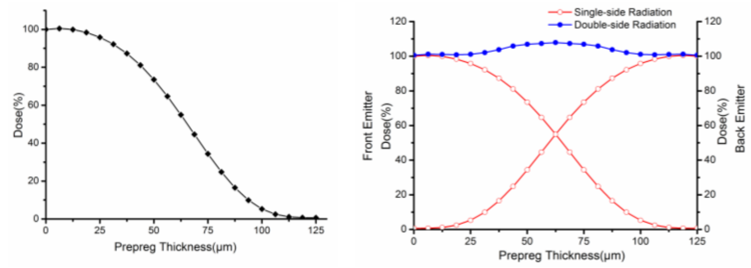


Figure 4.(a) Penetration curve of the 125keV E-Beam in prepregs; **(b)** Accumulated curve of the 125keV E-Beam in prepregs.

Fig.5 shows the relationship between the degree of cure of the prepreg vs. irradiation dose. An obvious increase is observed in the degree of cure when the dose increases over 50KGy and hardly increase above 90KGy. Hence, post thermal curing is essential to get the completely cured composites in low energy E-Beam curing process.

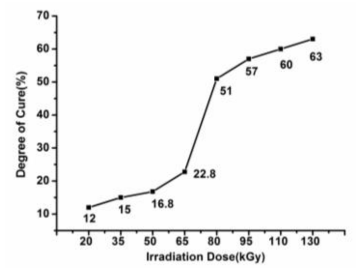
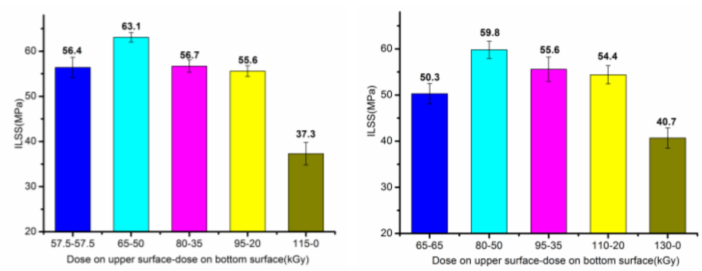


Figure 5. Relationship between the degree of cure of the prepreg vs. irradiation dose

Effect of irradiating method on ILSS

In this study, 21 laminates were fabricated with prepregs irradiated under a total dose(abbreviated to T-dose) of 115kGy, 130kGy, 145kGy and 160kGy. Under each T-dose, three irradiating methods were applied to the prepregs: the prepregs were single-side irradiated, double-side irradiated with equal dose on both sides and double-side irradiated with different doses on both sides. The U-dose means the received dose on the upper surface of the prepreg and the B-dose means the received dose on the bottom surface of the prepreg(seen in Fig.2).



(a)115kGy (b) 130kGy

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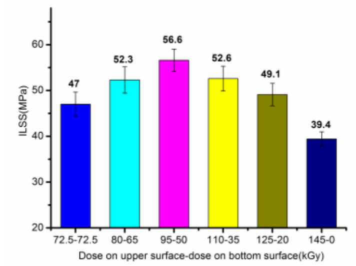
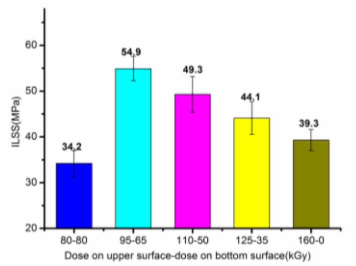


(c) 145kGy

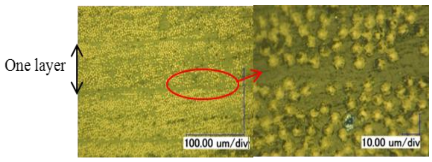


(d) 160kGy

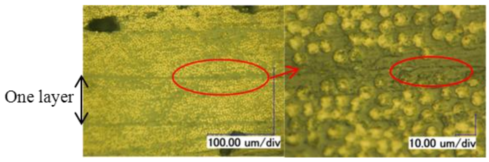
Figure 6. ILSS results of laminates fabricated with prepregs irradiated under different T-doses **(a)**115kGy;**(b)** 130kGy;**(c)** 145kGy;**(d)** 160kGy.

Fig.6 shows that the ILSS of laminates fabricated with prepregs single-sided irradiated are rather low. This mainly results from the fact that laminates fabricated with single-side irradiated prepregs cannot be fully cured after post-curing at 180 ℃ for 30 minutes. In the above-mentioned four laminates, although the dose on the upper surface of the prepregs is high to 160kGy, there is still no dose on the bottom surface of the prepreg owing to the poor transmission ability of the 125keV electron beam(Fig.4 (a)), which means that the bottom surface of the prepreg remains un-irradiated, leading to the poor ILSS of the laminates.

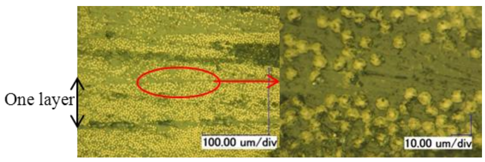
It can also be noted in Fig.6 that ILSS results of laminates with a dose difference on both sides of the prepregs generally exceed those with the same dose on both sides of the prepregs, respectively. A possible explanation for this is that E-Beam induced degree of cure on both sides of the irradiated prepreg is different, thus leading to different uncured resin content on the surfaces of the prepreg. In the preforming process, the bottom surface of the placing layer was adhered to the upper surface of the laid layer. During post-curing, the difference in uncured resin content contributed to cross-layer flow of resin between adhesive layers. The cross-section photographs typifying the adhering state between layers of the laminates were shown in Fig.7.



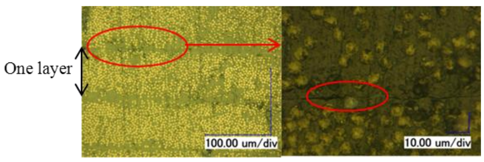
(a) 80-50kGy



(b) 65-65kGy



(c) 110-50kGy



(d) 80-80kGy

Figure 7. Cross-section photographs typifying the adhering state between layers of the laminates: **(a)** 80-50kGy; **(b)** 65-65kGy; **(c)** 110-50kGy; **(d)** 80-80kGy.

In this figure, laminates fabricated with different doses on both sides of the prepregs were observed to have better interlaminar quality and adhesive layers were closely combined. In comparison, microscopic slit appears between layers in the laminate irradiated with 65-65kGy and 80-80kGy. In addition to the physical union, crosslinking further strengths the interlaminar combination between adjacent layers during post-curing[27]. After irradiation, the free cations H^+ decomposed from the

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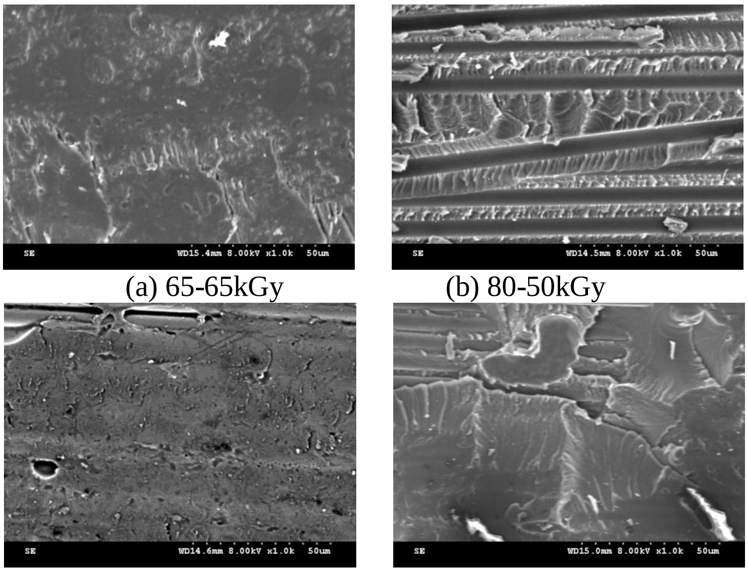
initiator and supposed to combine with the electronegative oxygen atoms of the epoxy groups. The content of decomposed H^+ increases within a certain irradiation range[28]. Different irradiation dose on both sides of the prepreg led to H^+ difference between adjacent layers, resulting in the phenomenon that H^+ on the surface that with more irradiation dose “attack” the oxygen atoms on the surface with less irradiation dose and crosslinking occurred between layers, which reinforced the interlaminar combination. Fig.9 shows microscope structures of the observed samples. It can also be seen in (b) and (d) that after ILSS tests, obvious ductile fracture trace is observed due to the large resistance when being damaged, further expressing the good adhesion and high ILSS. The SEM pictures show that laminates irradiated 65-65kGy (a) and 80-80kGy (c) have flat layered surfaces and damage occurs between the resin matrix, indicating that the laminates are easily to be damaged and layers are poorly adhered.



(a) 65-65kGy (b) 80-50kGy

(c) 80-80kGy (d) 110-50kGy

Figure 8. SEM photographs of fracture surfaces of laminates with a total dose of 130kGy and 160kGy(a) 65-65kGy;(b) 80-50kGy;(c) 80-80kGy;(d) 110-50kGy.

Although the fact that dose difference between adjacent layers improved the adhere quality owing to the uncured resin flow and cross linking between adjacent layers, ILSS results show that larger dose difference weakened the improving effect. In Fig.6, optimum dose difference was observed wherewith the best ILSS results were obtained under each T-dose.

3. Conclusions

In this paper, the low energy E-Beam in-situ curing technology in polymer-matrix composites was explored. Irradiation dose decreased significantly in the prepreg, inducing nonuniform dose distribution and degree of cure along the prepreg thickness direction. Three irradiation methods were applied to the prepregs. Results showed that laminates fabricated by prepregs single-sided irradiated were low in ILSS owing to the fact that single-sided irradiated prepregs cannot be completely cured at 180 ℃ for 30min. Laminates fabricated with prepregs with different doses on both sides have better interlaminar adhesion and ILSS compared to those fabricated with prepregs having the same dose on both sides.

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