Composites: Part A 65 (2014) 73–82

Contents lists available at ScienceDirect

Composites: Part A

journal homepage: www.elsevier.com/locate/compositesa

Low-energy electron beam cured tape placement for out-of-autoclave fabrication of advanced polymer composites

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ARTICLE INFO

Article history:
Received 2 December 2013
Received in revised form 3 April 2014
Accepted 8 June 2014
Available online 14 June 2014

Keywords:
A. Polymer–matrix composites (PMCs)
E. Autoclave
E. Cure
E. Lay-up (manual/automated)

ABSTRACT

The autoclave curing process for advanced polymer composites is labor and capital intensive, with the curing cost increasing dramatically by the growth in part size. In order to develop an out-of-autoclave (OOA) fabrication process for advanced polymer composites, a new process integrating automated tape placement with low-energy electron beam radiation curing was explored, by which in situ layer-wise curing of advanced composites can be achieved with the tape placement process. The irradiation process was optimized to get a homogenous curing, by tuning the electron beam dose-depth distribution in the prepreg material. Besides, the curing characteristics of the prepreg material by the low-energy electron beam irradiation was investigated and effect of exposure dose and post curing on curing degree, glass transition temperature (Tg) and interlaminar shear strength (ILSS) were characterized experimentally.

1. Introduction

Thermoset composite materials are used to a large extend today and automated thermoset tape placement machines are used to pre-form large-scale structures like wind turbines, ship structures and aerospace components. After pre-forming of the material, the complete mold structure is bagged and placed in an oven or autoclave to cure, depending on the desired performance. The automated placement of the material allows consistent part quality while the traditional autoclave based thermal curing method for thermoset composites faces critical challenges with the increasing of size and thickness of composite parts [1]:

- Long curing cycles with high energy consumption.
- Non-uniform curing of parts with large residual stresses due to the unavoidable temperature gradients.
- Large capital investment and operating expense.

Moreover, in many cases, an oven or an autoclave that is big enough for the composite part is not available or has limited availability. One good example is the "world's largest" autoclave (working area: φ 9 m × 25 m) developed by "ASC Process Systems" company to satisfy the curing demands from the Boeing-787's fuselage and wing skin carbon–fiber/epoxy parts. The size limitation of autoclave is a great challenge to large-scale advanced polymer composites, which is limiting increased use of advanced polymer composites in the wind and aerospace industry.

Researchers and industries have long desired to explore and develop low cost out-of-autoclave (OOA) composite fabrication methods and investigated different curing alternatives, including X-ray, γ-ray [2,3], ultraviolet [4–6], microwave [7,8], electron beam (EB) [9–11] etc. X-ray, γ-ray was proved to be especially suitable for thick composites (up to 300 mm) due to the extra-high electromagnetic energy [2,12]. However, the dose rates of X-ray and γ-ray are rather low compared with those of electron beams, resulting in the same curing cycle as autoclaves. Most importantly, curing by gamma ray and X-ray techniques involves great risk to widely apply these technologies for industry, considering the high level of radioactivity and the hardly disposable matter; UV curing is limited to open mold processes and transparent composites (mostly glass fiber reinforced) with limited thickness [13,14], due to the absorption behavior of UV radiation passing through the polymer and fiber materials. Microwave showed good potential for curing of pure polymer or glass/aramid fiber reinforced composites that have lower dielectric loss properties [15–17]. Research results of Boey et al. [18,19] showed that the stiffness and strength properties of the glass fiber composites cured by microwave are equivalent to those by autoclave curing. However, great challenges arise when microwave heating for carbon fiber composites using the traditional microwave ovens [7,20]: due to the high dielectric loss of the carbon fiber, the reflectance of the first few layers is

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http://dx.doi.org/10.1016/j.compositesa.2014.06.004
1359-835X/

too high to achieve efficient heating of thick laminates. Besides, the incorporation of high electrical conductive carbon fibers results in the formation of local hot spots and electrical arcing. Recently Feher and Meyer et al. [21,22] developed microwave autoclaves by tuning the electromagnetic frequency and heating field and successfully achieved accelerated heating of carbon fiber polymer composites, however the issues relating the energy efficiency, heating depth and homogeneity, local hot spots and possible hazards to the human body of microwave are still challenging aspects to widely apply microwave curing for advanced polymer composites in industry. The detailed overview of different curing methods, including the application status and existing challenges for advanced polymer composites is provided in the reference paper [23].

EB curing is based on accelerated electrons that can provide energy for the initiation of the curing process by the decomposition of the radiation-sensitive initiator. Since AEROSPATIALE company of France firstly investigated the EB curing of advanced composites for aerospace applications from the early 1970s and manufactured a missile fuel tank by EB curing [3], the application of EB curing for advanced polymer composites has obtained great development. Several airplane parts, including bulkhead and wing skin, were successfully manufactured [10,11]. EB induced radiation curing is a very attractive technique for the fabrication of high-performance composite materials and has been considered as a potential alternative for OOA curing of advanced polymer composites [24]. However, EB curing of thermoset composites after complete layup needs high-energy electrons (5–15 MeV) to penetrate the whole thickness of the part, which needs large capital costs for the high-energy EB emitters, the concrete walls and ceilings to shield x-rays which are produced as a by-product of the EB process. Besides, because of the non-uniform dose distribution of the electrons through the material and reaction exotherm generated inside, non-uniform curing, uneven mass shrinkage, void and bubble formation could occurs, which might lead to an improper consolidation of the fiber/resin composite material. If the composites are too thick and exotherm is high, severe thermal degradation of the matrix material may occur resulting in potentially hazardous processing conditions and a terminally damaged composite product. Gang et al. [25] investigated the effects of the electron beam dose and initiator concentration impacting factors on the curing homogeneity and pointed out that for thick resin products layer upon layer irradiation or irradiation from two sides could be more efficient to obtain a homogeneous crosslinking structure.

Aiming at the high operating and maintaining costs and poor curing homogeneity existed in the high-energy EB curing method, this paper explored a new low-energy EB cured tape placement process for OOA fabrication of thermoset composites. This process integrates automated tape placement method with a self-shielded low-energy (150 keV) EB irradiation unit for the on-line curing of the prepregs. The tape placement process provides significant consolidation pressure during the curing which is not available in the standard EB curing of composites, so enough consolidation and adhesion between layers can be expected simultaneously during the tape layup.

A low-energy EB cured tape placement system was set up and irradiation process was optimized to obtain a homogenous curing of the prepreg. The curing behavior of composites by low-energy EB irradiation was characterized, and the effect of processing parameters, including exposure dose and post-curing, on interlaminar shear strength (ILSS) was investigated experimentally. The layer-wise low-energy EB curing tape placement method in this paper provides a low-cost and efficient way to fabricate thermoset composite laminates without autoclave processing and it has a great industrial application potential for high performance composite areas. Besides, this method allows the fabrication of large structures with low energy consumption, more flexibility and simplicity compared to the large capital and tooling expenditures, poor curing homogeneity and size restrictions inherent in autoclave curing.

2. Experimental

2.1. Materials

EB curable prepregs were prepared with epoxy resin EB99-1 (Beijing Institute of Aeronautical Materials, China) with cationic initiator iodonium salt (Sartomer CD1012, USA) by a 1.5 phr concentration and carbon fiber T700 (TORAY, Japan) in Beijing Institute of Aeronautical Materials, China. The fabricated prepreg has a fiber surface density of 130 ± 5 g/m^2, resin content of $35 \pm 3\%$ and thickness of 0.125 mm. Thermal curing prepregs (USN12500) were provided from Weihai Guangwei Group, China.

2.2. Fabrication process of composites

Composite laminates were fabricated by the setup illustrated in Fig. 1. After being irradiated to a certain dose with low-energy EB emitters (Advanced Electron Beams, USA), the irradiated EB curable prepregs were immediately compacted on the mold surface by tape placement machine. The emitting energy and beam current parameters of the emitters are adjustable in the range 80–150 keV and 0–25 mA. The tape placement machine used for the experiment is developed in the State Key Lab for Manufacturing Systems Engineering, Xi'an Jiaotong University, China. The tape placement machine can place multiple prepregs simultaneously, with a maximum placement speed of 30 m/min and compaction force of 1200 N. The width of the EB curing tape that is used in the experiment is 100 mm; the length and diameter of the compaction roller of the tape placement head is 120 mm and 60 mm; the placement speed and compaction force used for the experiment is 5 m/min and 1100 ± 25 N.

2.3. EB exposure dose

The surface exposure dose of composites D (kGy) is calculated according to Eq. (1).

$$D = K \cdot I/S \tag{1}$$

where K (kGy m/min/mA) is the application efficiency coefficient of emitter, which is a combined factor of EB acceleration voltage, irradiation air gap, ambient gas species and emitter efficiency. I (mA) is the beam current, and S (m/min) is the irradiation speed of the prepreg.

The actual irradiated dose on the surface of the material was measured by dosimeter (B3000, GEX Corporation). After irradiation with EB, the dosimeters were immediately heat treated in

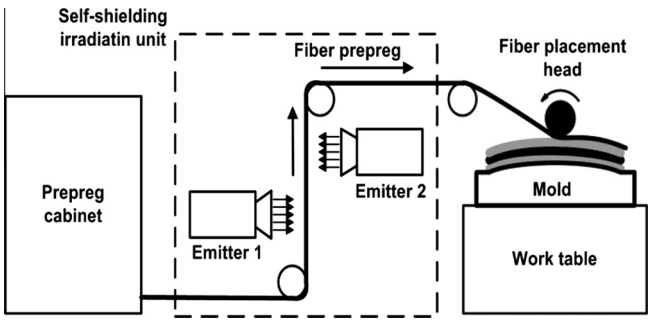

Fig. 1. Illustration of low-energy EB cured thermoset tape placement process.