Heat transfer studies for infrared radiation assisted curing in polymer composites

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Abstract: Conventional materials are replaced by polymer composites in every field due to its wide applications and its ease to custom make the required properties for the purpose it is designated for. Structural applications require higher strength, and this is achieved only by better curing of polymer composites during its processing. Elevated temperature curing helps to improve cross linking of polymers. In this regard infrared radiation curing is one of the methods adopted in the recent days for elevated temperature curing of polymer composites. It is one of the efficient ways of curing polymer matrix composites. In this paper Infrared radiation heat transfer studies of polymer composite is reviewed and compared its efficiency with the hot air or conventional curing method. Further modelling is carried out to prove the efficacy of the same.

1. Introduction

Fiber-reinforced composites have seen many applications in industries due to their mechanical properties which include high strength to weight and stiffness to weight ratios, corrosion resistance, impact/damage tolerance characteristics and wear properties. Aerospace domain, marine, transport and sporting goods are the industries which uses these composites. Fiber reinforced composites have their applications in structures also. Fiber-reinforced composites use resin, matrix and fibers as reinforcement. To achieve complete cure, strength or designated strength the FRP composite has to be cured for certain duration at elevated temperature depending on the type of resin and hardener. This is one of the most vital processes in the fabrication of FRP composites. In this work infrared heating is studied in comparison to thermal curing process.[1,2]

In recent days, great range of thermal heating processes for curing are in application which including laser, microwave, hot shoe, hot gas, flame, oven, induction, ultrasonic, resistance heating etc. and to manufacture composite products thermal curing process plays a dominant role in industries for curing. Here heating mechanisms can be categorized into radiation heating, convection & conduction heating, induction heating, ultrasonic heating, resistance heating and thermal additive-based heating [3,4].

2. Curing of Material

Fiber reinforced composite is a flexible material that can be prepared to fit the required design of various structural applications. The required properties of a fiber reinforced composites for a definite application needs a thorough and a better understanding of chemistry of curing and post curing reactions. The reactions are affected by the processing conditions, chemistry of the resin and the curing agent. A complex molecular network is generated in epoxy resin as a reaction between resin and curing agent, and the molecular characteristics influence the end-product performance directly.

The curing of epoxy resin by amine curing agents is expressed by the formula as shown in figure below; the active hydrogen in primary amine reacts with an epoxy group to form secondary amine, which further reacts with an epoxy group to cure. Then, the resultant tertiary amine polymerizes epoxy groups. Epoxy resins possess better chemical and corrosion resistance, has toughness and flexibility,

along with good mechanical and electrical behavior with outstanding adhesion to many different striates [5].

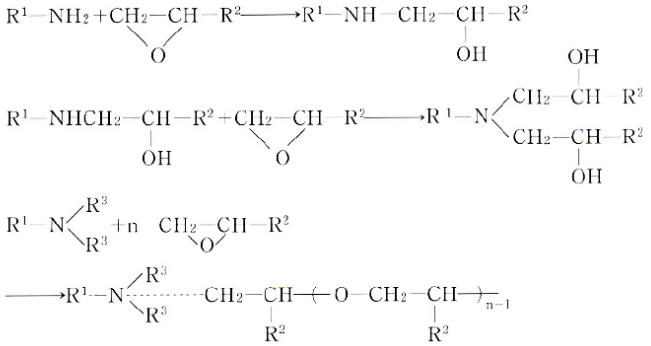


Figure 1. Epoxy resin curing reaction

Fiber reinforced polymer composites curing is one of the time consuming and critical step in achieving higher strength which is obtained by processing of polymer composite through cross linking of molecular chains. There are different stages of curing stage A, stage B and stage C curing. Stage A curing takes place at room temperature which is the initial stage initiated by curing agent or hardener. Stage B is the intermediate stage where the resin mixture is glassy and still in the uncured state which requires external heat for complete curing. Stage C takes place by further cross linking of the polymer molecules as it is exposed to higher temperatures or initial cure cycle temperature. This stage is necessary to achieve the required mechanical properties and to improve the bond strength. Required time for processing and curing of polymer composites differ according to the type of matrix material, hardeners and reinforcement selected along with the type of curing selected whether thermal or radiation method of cure. Ramping and soaking during curing at different temperatures, deciding the optimum cure cycle for the process selected is a prodigious task. Cure cycle for typical epoxy with hardener is shown below and the table indicates the temperature and time schedule.[6,7]

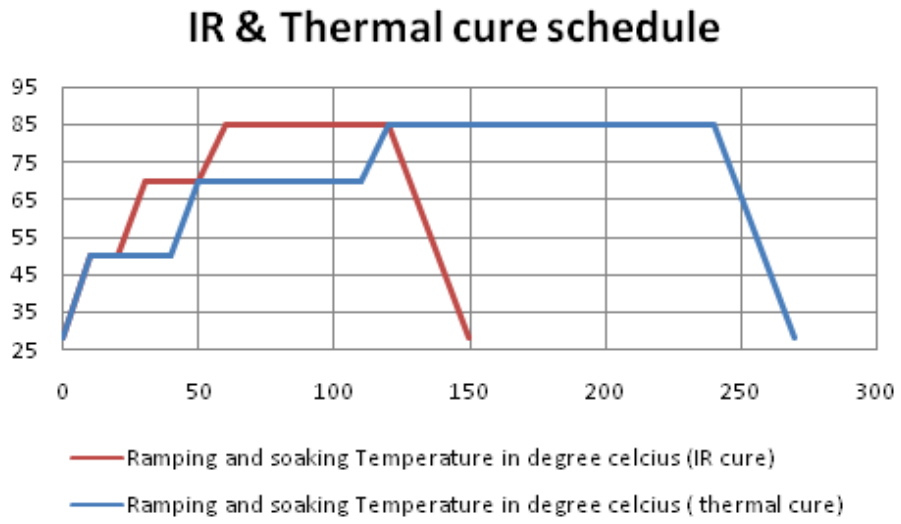


Figure 2. IR and Thermal cure schedule

Table 1

Temperature in ^{0}C	IR schedule, min	Thermal schedule, min
28-50	10	10

50	10	30
50-70	10	10
70	20	60
70-85	10	10
85	60	120
85-28	30	30

The temperature range where a thermosetting polymer changes from a "glassy" state to a more elastic, or "rubbery" state is the glass transition temperature (Tg). The Tg is determined by a number of factors like the chemical structure of the epoxy resin, type of hardener selected and the degree of cure. Glass transition temperature (Tg) explains the degree of cure of the composite[7]. Earlier researchers have proved that the Tg has a direct relationship with the mechanical properties of the composites. Many researchers have worked on Ultraviolet and Microwave methods but the work with respect to Infrared curing of composites has been carried out by very few researchers. IR can rapidly heat components and is ideally suited for flat surfaces. IR offers fast curing rate with precise control, high efficiency and minimal environmental impact. [8,10]

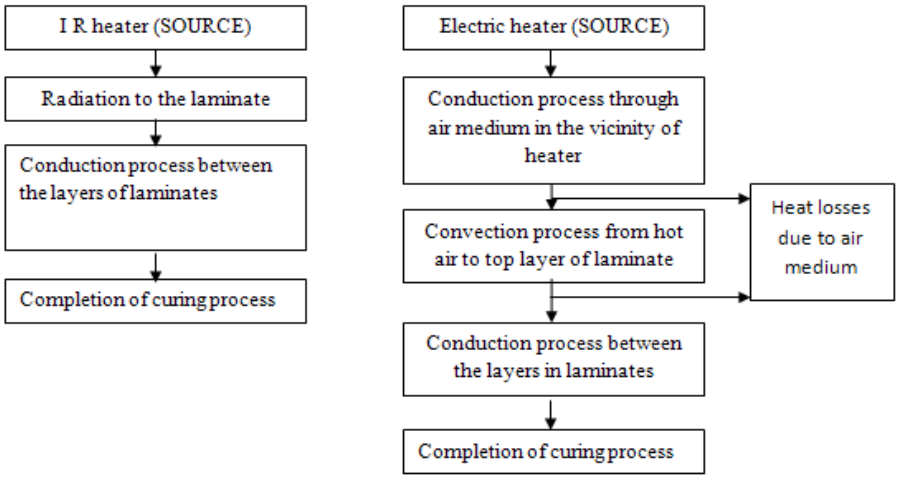
3. Radiation heat transfer and curing mechanism

This paper mainly focuses on heat transfer studies by Infrared Radiation curing in polymer composites. Radiant energy has gained increasing markets in polymer processing due to its low thermal conductivity. Most of the polymers which are around 0.1-0.6 W/m/K is been used. The main advantages of IR heating is the direct transfer of energy between the source and the component product that has to be treated, due to its high power density, control flexibility, low investment cost and adaptable, reduced curing time and reduced volatile emission[9].

Infrared light ranges between visible light and microwaves which is measured in microns on the electromagnetic spectrum. The wavelength of the IR emitter source is determined by its temperature, so by changing its temperature the peak wavelength can be controlled. All emitters which are used can be adjusted for required wavelength but not all heaters are designed to discharge the complete spectrum of different wavelengths like short, medium and long wavelengths. [11]

Curing by Infrared radiation is a different phenomenon compared to thermal curing. Here Infrared radiation is made to incident directly on the top surface of the composite laminate that is to be cured and the internal layers are heated by conduction of heat that flows from the top layer.

Flow chart 1 shown explains the mechanism of heat flow to the composite laminate by IR and hot air/thermal curing methods.



Flow chart 1

The radiative heat flux λ_1 emitted by infrared lamps be able to either get reflected (ρ_λ) , absorbed (α_λ) or transmitted (τ_λ) by the polymer as shown in fig. 3

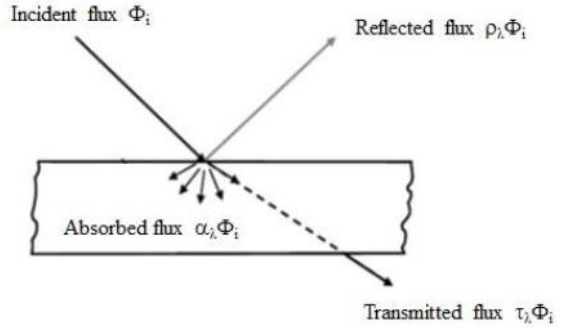


Figure 3. Radiative heat from infrared lamps

Electromagnetic wave is in charge of heat transfer by radiation travelling at the speed of light (c = 2.998x108 m/ s in vacuum), which is called electromagnetic radiation. The energy emitted is a kind photons which exhibits as electromagnetic wave and is characterized by wavelength λ and frequency ν:

$\lambda = c/\nu$ .....(1)

Thus, first Kirchhoff's law expresses the energy balance as

$\alpha_\lambda + \tau_\lambda + \rho_\lambda = 1$ .....(2)

In addition, using Kirchhoff's second law, we can assume the energy absorbed is equal to energy emitted:

$\varepsilon_\lambda = \alpha_\lambda = 1 - \tau_\lambda - \rho_\lambda$ ....(3)

Where ε_λ is the spectral emissivity coefficient. Diffuse reflection is characterized by a specific ratio σ/λ where σ is the Root Mean Square (RMS) roughness of the surface and λ the wavelength of the incident radiation.

If $\sigma/\lambda << 1$, then the surface is assumed to be a specular; and if $\sigma/\lambda >> 1$, the surface is assumed to be a diffuse one..[7]

The expressions for transmission and reflection factors from the literature survey are

$$T_\lambda = (1-\rho_\lambda)^2 \tau_\lambda / ((1-(\rho_\lambda \tau_\lambda)^2)$$

$$R_\lambda = \rho_\lambda [\, 1 + \tau^3_\lambda T_\lambda] \qquad \text{.....(4)}$$

Infrared emitters vary mostly because of the technology, their geometry and their temperature T_λ . The temperature plays a major because it affects the maximum power and wavelength range associated with the element. Infrared radiation heating is based on conversion of electromagnetic energy to heat by resonance vibration of molecules. Polymers contain many couplings such as CH, CH2, CH3 and CC and these molecules are vibrating at specific frequencies.

The transient temperature in composite can be evaluated by the Fourier's modal equation by assuming an instantaneous equilibrium temperature between the resin and the fibres at each time[12]:

$$\rho_c c_{p,c} \frac{\partial T}{\partial t} = \nabla(k_c \nabla T) + v_m \rho_m H_u \frac{\partial \alpha}{\partial t} \qquad \text{..... (5)}$$

Where H_u= ultimate heat of reaction, α = degree of curing, and the ρ_c = density, $c_{p,c}$ = heat capacity, k_c = thermal conductivity of the composite

Exothermal heat reaction gets generated by a chemical reaction of the resin Q_r when resin starts to cure which can be expressed as

$$Q_r = (1 - V_f)\rho H_r \frac{d\alpha}{dt} \quad \text{.....(6)}$$

Heat conduction in the material can be expressed by the equation given below

$$\rho C_p \frac{\partial T}{\partial t} = (k_x \frac{\partial^2 T}{\partial x^2} + k_y \frac{\partial^2 T}{\partial y^2} + k_z \frac{\partial^2 T}{\partial z^2}) + (Q_e + Q_r) \quad \text{.....(7)}$$

Here the epoxy resin curing process undergoes exothermic reaction during the high temperature curing process

The density ρ and the heat capacity c_p of the laminates can be easily calculated as the weighted averages

$$\rho_c = \rho_m \rho_f / (\rho_m w_f + \rho_f w_m) \quad \text{.....(8)}$$

$$c_{p,c} = c_{p,m} w_m + c_{p,f} w_f \quad \text{.....(9)}$$

$$k_c = k_m k_{f,t} / ((1-v_m) k_m + v_m k_{f,t}) \quad \text{.....(10)}$$

$$w_m = (v_m / \rho_f) / ((v_m / \rho_f) + ((1-v_m)/ \rho_m)) \quad \text{.....(11)}$$

$$w_f = 1 - w_m \quad \text{.....(12)}$$

Where $k_{f,t}$ = transverse thermal conductivity of the fibers, w = weight fraction, v = volume fraction, the subscript f stands for the fibers, and m for the matrix. The above equations can be solved at each time in order to calculate the distributions of temperature, degree of curing and time involved in it.

From the above equations and by mechanism of heat flow as depicted in flow chart it is clearly understood that IR radiation is better as there is negligible losses in processing of curing the laminates [13,14,15].

4. Materials and methods

Materials used for the preparation of fiber reinforced composite is bifunctional diglycidyl ether of bisphenol A (DGEBA) type epoxy resin with the curing agent triethylene tetramine TETA (HY951) with stoichiometric ratio of 100:11. The fiber reinforced laminate has fiber to resin ratio of 65:35, E glass fiber bidirectional of 300gsm is used to prepare the laminate by hand layup method and further cured by thermal and IR curing. The schedule of the curing is shown in the table 1.

5. Experimental setup

The experimental setup we have is a curing chamber is shown in Fig.4. The curing chamber is with the dimension of 400 × 400 × 400 mm and is made of stainless steel material. Infrared heater is mounted on the top surface of the chamber and thermal coil heater is mounted on the side walls of the chamber. The heat transfer to composite laminate is by convection and conduction in case of thermal curing by hot air. The air is drawn in to the chamber by the path provided inside the chamber and the same is being recirculated until the desired temperature of the laminate is achieved. The thermocouples are mounted at various points in the curing chamber to sense the ambient air temperature. This device controls the temperature by cutting off the power supply to one or more number of thermal heaters according to the set soaking or ramping schedule programmed by the user.

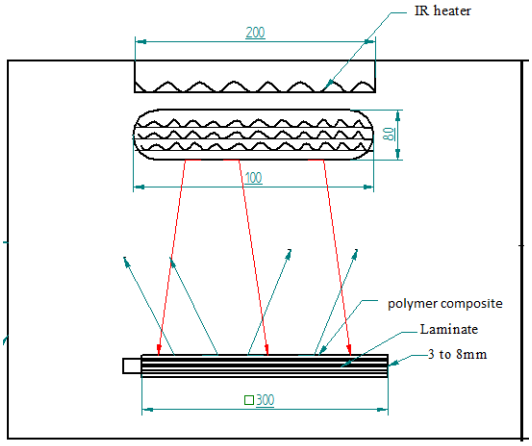


Figure 4. Experimental setup

6. IR Thermal image studies

Temperature distribution study was made through the IR thermal imaging camera FLIR C2; it is a high-sensitivity detector which captures fine differences in temperature and thermal patterns, this device forms a heat zone image using infrared radiation. As seen from the figure 5 & 6 IR radiation helps in uniform distribution of temperature when compared to thermal curing.

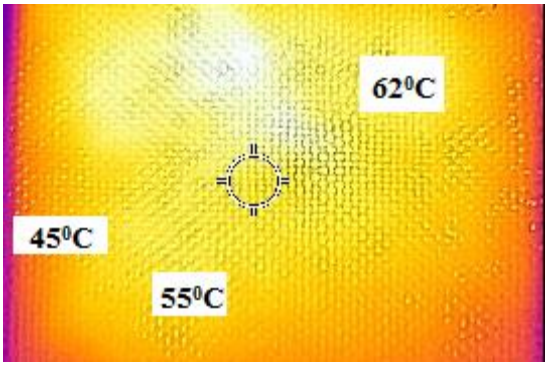


Figure 5. Non uniform temperature distribution in thermal curing

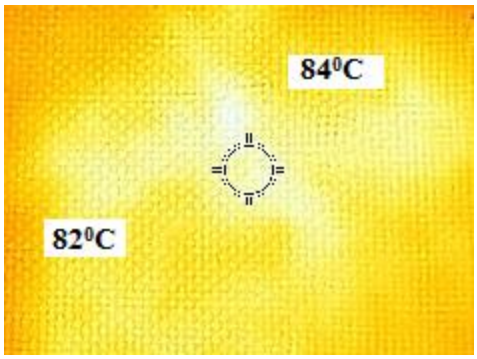


Figure 6. Uniform temperature distribution in IR curing

7. Results and conclusion

Tensile samples were prepared as per ASTM D638 standards and glass transition were determined by DSC thermal studies using ASTM D 3418 standards.

From the above test it indicates that tensile strength is high in case of IR curing due to better bonding between fiber layers and resin. Further strong dimensional structure is formed due to better cross linking of polymer and hence better resistance to the applied tensile load and leading to high tensile strength. Further higher Tg indicates uniform curing and better strength as higher Tg indicates improved inter laminar bonding and better cure compared to its counterpart (i.e Thermal curing). Matrix cracking has appeared only after application of half of the tensile load as assumed during the test. From the above results and mathematical modelling it is concluded that IR curing can be a better alternative and clean source as compared to other methods of curing for polymer composites.

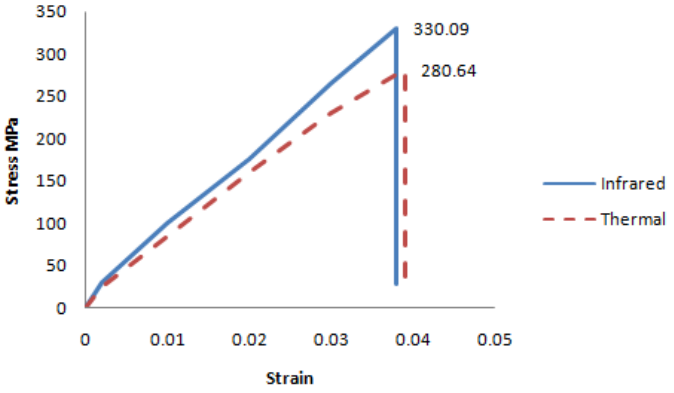


Figure 7. Tensile Strength

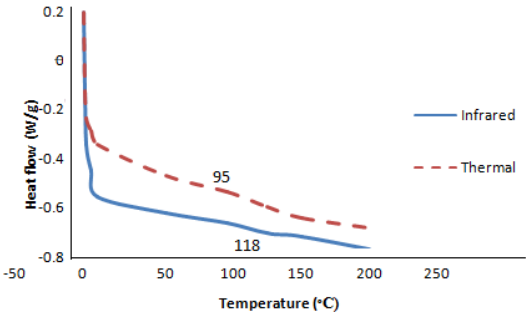


Figure 8. Thermogram

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