

A multi-pattern compensation method to ensure even temperature in composite materials during microwave curing process

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Abstract:

Microwave curing technologies have many advantages in manufacturing fiber reinforced polymer composite materials used in aerospace products, compared with traditional autoclave curing technologies. However, the uneven electromagnetic field of microwave in the cavity of the curing chamber results in uneven temperature on the surface of composite laminates during curing, which has been a major obstacle in industrial applications worldwide. Existing methods attempted to solve the problem by the random superposition of uneven electromagnetic fields, but the results were still not satisfactory to meet the high quality requirements of aerospace parts. This paper reveals the one-to-one correspondence between heating patterns of composite parts and microwave curing system settings, and reports a new concept to solve this problem by continuously monitoring and compensating the uneven temperature distribution in real-time. Experimental results from both fiber optical fluorescence sensors and infrared thermal imagers showed significant improvement in temperature uniformity compared with existing methods.

Key words: A. Polymer-matrix composites (PMCs); D. Process monitoring; E. Out of autoclave processing; E. Cure.

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1. Introduction

Fiber reinforced polymer composites with strong mechanical properties are increasingly used in aerospace products [1, 2]. According to an investigation recently carried out in collaboration with Chinese Aircraft Industrial (Group) Co., around 98% composites used in the aerospace industry are fabricated using autoclave curing technologies, where the material is placed in a chamber and heated by the circulating airflow [3]. However, the technology has a number of problems which restrict further improvement of product quality and manufacturing efficiency [4]. For aerospace applications, the most important problem is the serious deformation of composites of large size with varying thickness, due to the large temperature gradient in the thickness direction. Other problems include long process cycles and high energy consumption [5]. For example, the annual output of the A350XWB will be 156 airplanes after 2018 [6], thus 312 wings need to be manufactured. For some composite parts, only the curing process may take up to 24 hours with a maximum energy consumption of 4070KW per hour for an autoclave of size $\Phi 5\text{m} \times 14\text{m}$ [7, 8], and the part deformation can be very severe [9]. This cannot meet the increasing demands for large quantity of high performance composites in modern aircrafts.

As an alternative to traditional autoclave curing technologies, microwave curing technologies can reduce curing time and energy consumption, and also reduce the temperature gradient within the composite material during curing. This is because microwaves can heat the whole volume of the material at the same time [10], thus greatly reducing the deformation of composite parts and improving the efficiency of the curing process [11]. To date, a lot of research work had been conducted on microwave curing of composites materials, including fundamental principles [12], curing kinetics [13], fiber/matrix interfaces [14], reducing temperature gradient [15] and mechanical properties [16, 17].

However, microwave curing technologies have not been widely applied in the aerospace industry because of the difficulties in ensuring an even temperature on the

surface of composite laminates during curing [18, 19]. The uneven temperature distribution is caused by the uneven resonance of the electromagnetic field in the cavity of microwave ovens [20]. Resonance can be considered as the effect where waves are incident from several directions at the same time. For any arbitrary point in the cavity, the separate wave fields incident from different directions interfere each other, i.e., they combine constructively and destructively in an alternating pattern, and form a standing wave during the superimposition [21]. Over time ‘hot spots’ and ‘cold spots’ (relative to the hot spots), corresponding to antinodes and nodes of the standing wave will inevitably appear on the surface of composite materials, leading to the uneven in-plane temperature distribution. Because composite materials are basically laminated plate structures, the uneven temperature distribution on their surfaces has a significant impact on their curing performance, which can directly contribute to severe warpage and even local ablation or under-treatment.

In order to solve the uneven temperature problem, different ways had been attempted in the past which can be classified into four categories. The first one is to focus on the design of the shape and size of the microwave cavity [22]. For example, the uniformity of the microwave field can be improved by increasing the size of the cavity. This is because the number of resonant modes (the distribution state of the electromagnetic field) within a microwave applicator increases rapidly as the size of the cavity increases, and sometimes the different resonant modes within the applicator are possible to have complementary effects. The second way is to use multiple microwave sources within the cavity since the resonant modes associated with different sources are able to overlap, which may further enhance the heating uniformity [23]. The third way is to generate a relative movement between the material and the electromagnetic field [24]. An example can be found in a microwave oven at home which is often equipped with a turning table that rotates the plate with food during operation. The purpose of the turning table is to reduce the effect of multiple hot spots by moving the object being heated through areas of high and low power fields alternately, so as to achieve uniformity in temperature of the food. The fourth way is to adopt variable-frequency

microwave systems for materials processing, which can generate many different resonant modes by repeatedly applying different microwave frequencies thus achieving uniformity of power within the microwave cavity [25, 26].

The above existing methods have, to different extents, improved the uniformity of microwave heating by random superposition of the uneven electromagnetic field within the microwave cavity. However, these methods cannot solve the problem from the scientific point of view, and the uneven temperature problem during microwave curing remains as a major challenge in the manufacturing of advanced composite materials [18, 19]. This paper reveals the relationship between heating patterns of composite parts and microwave curing system settings. On this basis, a multi-pattern compensation method is proposed to achieve better uniformity of temperature on the surface of composite laminates during microwave curing. This method, through monitoring the uneven temperature distribution and applying appropriate compensating HPs in real-time, can significantly improve the homogeneity of the temperature field of composite parts during curing.

2. Idea of the multi-pattern compensation method

Through extensive experimental research, the authors found that there is a one-to-one correspondence between heating patterns (HPs) of composite parts and microwave curing system settings (MCSSs), as illustrated in Fig.1. Corresponding theoretical analysis is presented in Section 5.1. Here, HP are defined as the distribution law of the microwave power on the composite surface, which can be mathematically expressed as a matrix which contains the information of the microwave power and position.

$$\mathbf{HP} = \begin{bmatrix} P_{11} & P_{12} & \dots & P_{1n} \\ P_{21} & P_{22} & \dots & P_{2n} \\ \vdots & \vdots & \ddots & \vdots \\ P_{m1} & P_{m2} & \dots & P_{mn} \end{bmatrix} \quad (1)$$

where P_{mn} is the microwave power at a certain point on the composite surface. The

MCSS represents a couple of parameters regarding the resonant applicator, the microwave input and the composite part. Among them, parameters of the composite part (material, ply, shape, size and position) and the microwave equipment (shape, size, filling medium of the applicator, and frequency of the microwave input) can be regarded as constants and are difficult to be adjusted during curing, when a certain composite part and a certain industrial microwave oven (often with multiple microwave magnetrons) are selected. Fortunately, the position and the number of microwave inputs, as well as the power ratio between them can be easily controlled by adjusting the switches of various magnetrons of the oven as an electronic process. In this paper, the position and number of microwave inputs were used as the control strategy of the MCSS, and can be expressed mathematically as a vector.

$$\text{MCSS} = [\delta_1, \delta_2, \dots, \delta_l] \quad (2)$$

where δ_l is the switch state of the l th microwave input, which is a binary number and can be valued at 0 or 1. As mentioned above, the heating pattern of the composite can be controlled by adjusting the control strategy of the MCSS.

$$f(\text{MCSS}) = \text{HP} \quad (3)$$

According to the above analysis, the HP of the object being heated will not change as long as the related MCSS remains constant. Hence, when a part (or a new one of the same) is heated for a new run, the HPs collected beforehand can be used as a useful database to adjust its uneven temperature distribution. More specifically, when a certain temperature distribution is monitored, a HP with a complementary heating preference would be most beneficial to realize a uniform in-plane temperature distribution, especially when the high/low power sections of the HP are cold/hot spots for the current temperature distribution (see Fig.2). This is the idea of the multi-pattern compensation method. It overcomes the limitation of random superposition principle in traditional method, and use complementary HPs to ensure even curing temperature during the whole curing process.

3. Implementation of the multi-pattern compensation method

A process control system is developed to implement the multi-pattern compensation method. As shown in Fig.3, the structure of the system can be divided into two parts. One is aimed at improving the temperature uniformity of the composite part, and the other is to keep the average temperature following the setting temperature. It can be seen that before a composite part to be cured its database of HPs needs to be constructed. When the curing process is started, the temperature distribution of the part is monitored and analyzed in real time. If the maximum temperature difference exceeds a preset threshold ΔT_{max} (e.g., 6°C), the HP selection controller will rapidly search the database for a HP that would alleviate the temperature heterogeneity the most. Once the HP is selected, the computer will rapidly adjust the switches of the magnetrons of the oven according to the related control strategy. Since composites are often cured by a fixed temperature process, the input power of these magnetrons also needs to be adjusted by a PID power controller to keep the average temperature following the setting temperature.

3.1 Strategy of HP database construction

As mentioned above, a database of HPs for a composite part needs to be set up before it is to be cured by microwave. This is accomplished by a preheating process of the part. The details are as follows.

(1) Equivalent expression of HP. Since the microwave power distribution on the composite surface is unmeasurable without disturbing the original microwave field, another physical quantity that is directly proportional to the microwave power has to be used to solve this problem. During microwave curing, the heat transfer model within the composite laminate can be described as below [27]:

$$\rho c_p \frac{\partial T}{\partial t} = Q_e + Q_c + Q_a \quad (4)$$

$$Q_e = 2\pi f \varepsilon'' E^2 \quad (5)$$

$$Q_c = k_x \frac{\partial^2 T}{\partial x^2} + k_y \frac{\partial^2 T}{\partial y^2} + k_z \frac{\partial^2 T}{\partial z^2} \quad (6)$$

$$Q_\alpha = \rho H_R \frac{\partial \alpha}{\partial t} \quad (7)$$

where ρ refers to the density of the composite, c_p is the specific heat of the composite, t is the time, T is the transient temperature filed at time t , Q_e is the absorbed microwave power, Q_c is the heat transfer within the composite, Q_α is the exothermic heat of cure reaction, f is the frequency of the microwave, ε'' is the dielectric loss of the composite, E is the electric field intensity, k_x , k_y , k_z are the thermal conductivity along the x , y , z direction, H_R is the amount of heat release when the resin is fully cured, α is the degree of cure. In order not to affect the final performance of the product, the preheating process is conducted below the curing temperature of the composite, so it is unnecessary to consider the effect of the exothermic reaction. In the microwave curing process, microwave propagates at the speed of light and rapidly reaches the surface of the material, causing instantly sharp rise in temperature, then the heat transfer phenomenon just occurs with a much slower conduction rate between the relatively hot and cold areas. Hence, the impact of Q_c is very small compared with that of Q_e , and it can be approximated to a constant or even ignored during the preheating process, especially when a short heating time is adopted for each control strategy. According to Eq.4, the heating rate of each point is linear with the microwave power at that point on the composite surface, so it was used to describe the HP of the composite in this paper.

$$\mathbf{HP} = \begin{bmatrix} P_{11} & P_{12} & P_{1n} \\ P_{21} & P_{22} & P_{2n} \\ P_{m1} & P_{m2} & P_{mn} \end{bmatrix} \propto \begin{bmatrix} \frac{dT_{11}}{dt} & \frac{dT_{12}}{dt} & \frac{dT_{1n}}{dt} \\ \frac{dT_{21}}{dt} & \frac{dT_{22}}{dt} & \frac{dT_{2n}}{dt} \\ \frac{dT_{m1}}{dt} & \frac{dT_{m2}}{dt} & \frac{dT_{mn}}{dt} \end{bmatrix} \quad (8)$$

During the multi-pattern compensation process, although the dielectric properties of the composite may change with the progress of the curing reaction [28], it will only affect the specific value of heating rate at each point according to Eq.4 and 5, but the heating preference/pattern will almost not change since the composite laminate is uniformly heated (compensation will be triggered when temperature difference exceeds 6°C) from the beginning to the end, thus the dielectric properties of each point is uniformly changed. As for the exothermic heat of cure reaction, it will influence the temperature distribution of the composite during microwave curing, but the temperature distribution is monitored in real-time by temperature sensors, and then a normal compensation process will be carried out. That is, the reaction induced HPs are eliminated by temperature measurement, and it has nothing to do with the HP compensation process. Therefore, the heating pattern database established during the preheating process can be used for the whole curing process and no correction patterns are needed in general during the HP compensation process.

(2) Number of HPs needed to be extracted. For a certain microwave cavity, the diameter of most hot/cold spots without considering heat transfer can be estimated as follows, because there are two cold spots and hot spots in each wavelength range:

$$D = \frac{c}{4f_r} \quad (9)$$

where c is the speed of light, f_r is the resonant frequency of the microwave cavity which is around the frequency of the microwave input. Thus, at least n temperature measuring points are needed to effectively reflect the temperature distribution of the composite surface. As shown in Fig.4, if only one spot is considered during the compensation process, n HPs are needed to construct the HP database, making this spot, for example hot spot or cold spot, of these HPs spread over the whole surface of the composite part.

$$n = \frac{A}{0.25\pi D^2} \quad (10)$$

where A is the surface area of the composite part. If two spots are considered during

the compensation process, $n(n-1)$ HPs are needed to construct the HP database, making the two spots of these HPs spread over the whole surface of the composite part. Similarly, if i spots are considered during the compensation process, N HPs are needed to construct the HP database.

$$N = \frac{n!}{(n-i)!} \quad (11)$$

(3) Collection of HPs. The composite part is preheated under a variety of (about $15N$ according to the experience) control strategies (i.e. different combination of various microwave inputs), thus a large number of HPs would be obtained, from which the HPs mentioned above can be extracted to construct the HP database. As shown in Fig.5, If the value of N is relatively small, HPs can be obtained manually by a cyclic heating-cooling treatment method (Only one HP is obtained in one cycle). If not, it is better to make the control strategy programmed so that the computer can continuously execute multiple times in a single cycle for time saving. Further in-depth research on the HP collection process is to be conducted in the authors' on-going research work.

3.2 Strategy of uneven temperature compensation

During the microwave curing process, the uneven temperature of the composite part is compensated based on the established HP database. In this paper, only the hottest spot or the coldest spot was taken into account during one compensation step, considering the compensation accuracy and efficiency at the same time. When the curing process is started, the temperature distribution on the surface of the composite part is monitored in real time. If the maximum temperature difference exceeds a preset limit, a judgement will be carried out. If $|\bar{T} - T_{\max}| \geq |\bar{T} - T_{\min}|$ (\bar{T} , T_{\max} and T_{\min} are the average, maximum and minimum of the measured temperature), the hottest spot on the part will be compensated preferentially. Specifically, the computer will search the HP database very quickly for an appropriate HP which has the lowest heating rate at the corresponding position. Else, the coldest spot on the part needs to be compensated

preferentially. After the appropriate HP is found, the related control strategy will be applied to adjust the microwave field in the next moment by using data acquisition cards and a LabView interface. These monitoring and compensating steps are repeated until the composite part is completely cured. Of course, the compensation accuracy can be further improved by compensating the hottest spot and the coldest spot, even the secondary hot spot and the secondary cold spot et al, at the same time.

3.3 Strategy of power control

The purpose of the power controller is to keep the measured average temperature tracking the setting temperature. After the new control strategy (position and number of microwave inputs) is determined, the classical proportional-integral-derivative (PID) algorithm is adopted to control the microwave power [29]:

$$P(t) = K_p[e(t) + \frac{1}{T_i} \int_0^t e(t)dt + T_d \frac{de(t)}{dt}] \quad (12)$$

where $P(t)$ is the total power of current microwave inputs, $e(t)$ is the error between the setting temperature and measured maximum temperature, K_p , T_i and T_d are the proportional gain, integral factor and differential coefficient, respectively. When the total microwave power is determined, the power variation will be equally distributed to the current microwave inputs. This is because the same increment to the power of multiple microwave inputs will only lead to the variation in the specific value of heating rate at each point according to Eq.4 and 5, but the heating preference/pattern will almost not change since the composite laminate is uniformly heated (compensation will be triggered when temperature difference exceeds 6°C) from the beginning to the end, thus the properties of each point is uniformly changed. As a simple example, considering the situation of the microwave heating process with four inputs (see Fig.6), assume that the area marked 1, 2, 3 and 4 are mainly affected by the input of 1, 2, 3 and 4, respectively. When the power of these inputs is increased by Δp simultaneously, the heating rate of each area will increase by C (constant), but the HP of the composite (relationship

between the heating rates in these areas) will not be changed.

4. The experiment carried out

A 2.45GHz, 20KW microwave curing system was designed and manufactured by the research team, as shown in Fig.7 (a). Sixteen microwave sources are symmetrically distributed on each face of the octahedron oven. A fiber optical fluorescence measurement system, an infrared thermal imager (FLIR A300, see Fig.7 (b)), a LabView control module and a data storage/processing module were integrated into the equipment. A short carbon fiber (T300) reinforced epoxy composite plate of size 200mm×200mm×2mm, with the matrix volume fraction of about 40%, was prepared to validate the presented multi-pattern compensation method (see Fig.7 (c)). The curing schedule of this material was set at 90°C for 30min and 120°C for 60min with a heating rate of 3°C/min. Aluminium foils were stuck on the edge of the composite plate to avoid the arcing of carbon fibers under microwave irradiation.

The composite plate was first preheated by various control strategies to construct its HP database. In this work, the diameter of most hot/cold spots is about 5.4mm under the effect of both microwave and heat transfer. According to Eq.10, at least 17 temperature measuring points are needed to effectively reflect the temperature distribution of the composite surface. As a consequence, 20 fiber optical fluorescence sensors were uniformly distributed over the composite surface. The commonly used infrared thermal imager was given up due to the limitation of the vacuum packaging materials around the composite. Since this work only considers the hottest spot or the coldest spot during one compensation step, 40 HPs needs to be extracted to construct the HP database, in which 20 HPs were used to make the hottest spot of them uniformly distribute over the whole surface of the composite plate, and another 20 HPs were used to make the coldest spot of them uniformly distribute over the whole surface of the composite plate. A computer driven HP collection process was adopted. During the preheating process, the switch time of various control strategies was set as 4 seconds, the initial temperature was 25°C, the ramp rate was about 3°C/min,

and the temperature limit was set at 60°C because the curing reaction of the resin will be started when the temperature exceeds 60°C. Thus, about 175 HPs were recorded for one heating and cooling cycle. Subsequently, the 40 HPs mentioned above were extracted from collected 568 HPs.

After the HP database was constructed, the curing process of the composite plate was started. The preset threshold for compensation was set as 6°C, and the operating time of each HP is 8s. For comparison, three experiments were conducted. The first experiment adopted a traditional single pattern heating method, just proportionately increasing the microwave power to make the measured temperature following the process profile. The second experiment randomly changed the microwave power of all magnetrons of the oven to generate relative movement between the electromagnetic field and the composite plate, like the commonly used mode agitator or turntable. The third experiment used the proposed multi-pattern compensation method. For every experiment, three samples were tested.

5. Results and discussions

Based on the authors' experimental findings, this paper presents a multi-pattern compensation method to realize the uniform in-plane temperature distribution of composite parts during microwave curing. In order to ensure the feasibility of this method in principle, the theoretical analysis of the findings is first discussed in this section. Then, the effectiveness of the proposed multi-pattern compensating method is investigated in detail.

5.1 Theoretical analyses of the experimental findings

Since the HP of a composite part is directly affected by the electromagnetic fields inside a microwave oven, the influence of MCSSs on the electromagnetic fields inside the microwave oven is systematically discussed in this section. Taking a rectangular microwave oven for an example (see Fig.8 (a) and (b)), resonant frequencies of various electromagnetic modes inside the cavity can be determined as [12]:

$$f_{mnl} = \frac{1}{2\sqrt{\mu\varepsilon}} \sqrt{\left(\frac{m}{a}\right)^2 + \left(\frac{n}{b}\right)^2 + \left(\frac{l}{d}\right)^2} \quad (13)$$

$$\lambda_g = \frac{2\pi}{\sqrt{(\omega/c)^2 - [(m\pi/a)^2 + (n\pi/b)^2 + (l\pi/d)^2]}} \quad (14)$$

where f_{mnl} is the resonant frequency of TE_{mnl} and TM_{mnl} modes; μ and ε are the permeability and permittivity of the filling medium; a , b and d are the dimensions of the cavity in the x , y , and z directions which should meet the requirements of $a = m \frac{\lambda_g}{2}$, $b = n \frac{\lambda_g}{2}$, $d = l \frac{\lambda_g}{2}$; m , n and l are integers which determine the field type of electromagnetic modes; λ_g is the operating wavelength of electromagnetic modes; c is the speed of light; ω is the angular frequency of the microwave. It can be found from these relationships that the frequency of microwave sources (ω in Eq.14), the dimensions and filling medium of microwave cavities (a , b , d , μ and ε in Eq.13) are closely associated with the electromagnetic modes.

The influence of the applicator shape and heated object on the resonant frequency of the microwave cavity can be investigated by the perturbation method. As shown in Fig.8 (c) and (d), \mathbf{E}_0 , \mathbf{H}_0 and ω_0 are the fields and resonant frequency of the cavity before shape perturbation; \mathbf{E} , \mathbf{H} and ω are the fields and resonant frequency of the cavity after shape perturbation. For these two cases, the Maxwell's curl equations can be expressed as [27]:

$$\nabla \times \mathbf{E}_0 = -j\omega_0\mu\mathbf{H}_0 \quad (15a)$$

$$\nabla \times \mathbf{H}_0 = j\omega_0\varepsilon\mathbf{E}_0 \quad (15b)$$

$$\nabla \times \mathbf{E} = -j\omega\mu\mathbf{H} \quad (16a)$$

$$\nabla \times \mathbf{H} = j\omega\varepsilon\mathbf{E} \quad (16b)$$

Multiply the conjugation of Eq.15a by \mathbf{H} , and multiply Eq.16b by the conjugation of \mathbf{E}_0 , then:

$$\mathbf{H} \cdot \nabla \times \mathbf{E}_0^* = j\omega_0\mu\mathbf{H} \cdot \mathbf{H}_0^* \quad (17)$$

$$\mathbf{E}_0^* \cdot \nabla \times \mathbf{H} = j\omega \varepsilon \mathbf{E}_0^* \cdot \mathbf{E} \quad (18)$$

Applying the vector identity $\nabla \cdot (\mathbf{A} \times \mathbf{B}) = \mathbf{B} \cdot \nabla \times \mathbf{A} - \mathbf{A} \cdot \nabla \times \mathbf{B}$ to Eq.17 and 18 gives:

$$\nabla \cdot (\mathbf{E}_0^* \times \mathbf{H}) = j\omega_0 \mu \mathbf{H} \cdot \mathbf{H}_0^* - j\omega \varepsilon \mathbf{E}_0^* \cdot \mathbf{E} \quad (19a)$$

Multiply the conjugation of Eq.15b by \mathbf{E} , multiply Eq.16a by the conjugation of \mathbf{H}_0 , and do the same for these two formulas. Thus:

$$\nabla \cdot (\mathbf{E} \times \mathbf{H}_0^*) = -j\omega \mu \mathbf{H}_0^* \cdot \mathbf{H} + j\omega_0 \varepsilon \mathbf{E} \cdot \mathbf{E}_0^* \quad (19b)$$

Calculating the integration of the sum of Eq.19a and 19b on volume V , it gives:

$$\begin{aligned} & \int_V \nabla \cdot (\mathbf{E} \times \mathbf{H}_0^* + \mathbf{E}_0^* \times \mathbf{H}) dv \\ &= \int_S (\mathbf{E} \times \mathbf{H}_0^* + \mathbf{E}_0^* \times \mathbf{H}) \cdot \mathbf{ds} \\ &= -j(\omega - \omega_0) \int_V (\varepsilon \mathbf{E} \cdot \mathbf{E}_0^* + \mu \mathbf{H} \cdot \mathbf{H}_0^*) dv \end{aligned} \quad (20)$$

Because $\mathbf{n} \times \mathbf{E} = \mathbf{0}$ on surface S , $\mathbf{n} \times \mathbf{E}_0 = \mathbf{0}$ on surface S_0 and $S = S_0 - \Delta S$, the Eq.20 can be simplified as:

$$\omega - \omega_0 = \frac{-j \int_{\Delta S} \mathbf{E}_0^* \times \mathbf{H} \cdot \mathbf{ds}}{\int_V (\varepsilon \mathbf{E} \cdot \mathbf{E}_0^* + \mu \mathbf{H} \cdot \mathbf{H}_0^*) dv} \quad (21)$$

Similarly, the effect of the heated object can also be calculated by the perturbation method. As shown in Fig.8 (e) and (f), the resonant frequency of the microwave cavity after perturbation can be computed as follows.

$$\frac{\omega - \omega_0}{\omega} = \frac{-\int_{\Delta V} [(\varepsilon - \varepsilon_0) \mathbf{E} \cdot \mathbf{E}_0^* + (\mu - \mu_0) \mathbf{H} \cdot \mathbf{H}_0^*] dv}{\int_{\Delta V} (\varepsilon_0 \mathbf{E} \cdot \mathbf{E}_0^* + \mu_0 \mathbf{H} \cdot \mathbf{H}_0^*) dv} \quad (22)$$

It can be seen that the shape of the cavity (ΔS in Eq.21), the material, ply, shape and size of the heated object (ε , μ , \mathbf{E} , \mathbf{H} and ΔV in Eq.22) are also important factors to the resonance state in microwave applicators.

Overall, the electromagnetic modes inside the resonant applicator are confirmed when the above parameters are determined. However, the position and number of microwave inputs can affect the distribution of these modes inside the applicator, and

the power ratio between microwave inputs can influence the intensity of each mode. For example, the TE_{10l} mode induced by each microwave input in the rectangular oven of Fig. 8(a) can be expressed as:

$$E_y = \frac{-j\omega\mu a}{\pi} A_{10l} \sin \frac{\pi x}{a} (e^{-j\beta z} - e^{j\beta z}) \quad (23)$$

$$H_x = \frac{j\beta a}{\pi} A_{10l} \sin \frac{\pi x}{a} (e^{-j\beta z} + e^{j\beta z}) \quad (24)$$

$$H_z = A_{10l} \cos \frac{\pi x}{a} (e^{-j\beta z} - e^{j\beta z}) \quad (25)$$

$$E_x = E_z = H_y = 0 \quad (26)$$

where A_{10l} is an amplitude constant, β is the propagation constant of microwave. Since the coordinate in the above equations is a local coordinate system, the eventual field of the TE_{10l} mode is closely related to the superimposition of the field induced by various microwave inputs. Thus, the position and number of microwave inputs, as well as the power ratio between them have significant influence on the distribution of electromagnetic fields in the cavity. Additionally, the HP of the heated object will be directly affected by its position in the microwave applicator when the electromagnetic field is determined.

As mentioned above, all the parameters affecting the electromagnetic field inside the microwave oven are discussed theoretically. This further confirms the authors' experimental findings of the relationship between composite HPs and MCSSs.

5.2 Validation of the proposed multi-pattern compensating method

The temperature distribution on the composite surface under the traditional single pattern heating, random field variation heating and multi-pattern compensation heating was compared by the shown maximum, average and minimum temperature profiles (see Fig.9). Obviously, the temperature distribution was uneven during the traditional single pattern heating, especially at the beginning of dwell stages where the maximum temperature difference was about 34.3°C. As the dwell progressed, the maximum

temperature difference dropped to some extent because of heat conduction inside the material. In this experiment, it was found that the heating preference of the HP almost unchanged during microwave heating, which was demonstrated by monitoring the single pattern heating process of another sample without vacuum packaging by using the infrared thermal imager. Corresponding results are shown in Fig.10. The movement of the highest temperature between several hot spots may be attributed to the exothermic reaction of the resin matrix.

Fig.9 (b) presents a more uniform heating process through randomly adjusting the power of various microwave sources to provide variable microwave field, like the existing mode agitator or turntable. As shown, the maximum temperature difference was reduced to 26.4°C at the beginning of the second dwell stage. This was benefited from the randomly complementary effect of the extensive HPs during electromagnetic field variation. This may be demonstrated by the infrared thermal image in Fig.11. It can be observed that, unlike the traditional single pattern heating, the heating preference of the composite changes all the time. Sometimes good results can be obtained, and sometimes the situation is just the opposite. Overall, the uniformity of the temperature distribution is improved more or less, compared with that in single pattern heating.

The experimental results of the multi-pattern compensation heating are depicted in Fig.9 (c). It can be seen that the temperature difference was significantly smaller than both traditional single pattern heating and random field variation heating, throughout the whole curing process. The maximum temperature difference was only 11.2°C, which brings a reduction of about 67% and 58% compared with the traditional single pattern heating and random field variation heating. As shown in Fig.12, the heating preference of the composite also changes all the time in the whole multi-pattern compensation curing process. The temperature uniformity is greatly improved by using the complementary HP to compensate the monitored uneven temperature in real time.

The maximum temperature difference of the three heating processes was summarized in Fig.13. For the traditional single pattern heating, the maximum temperature difference increased rapidly in a linear mode during the heating process,

and gradually stabilized at the dwell stage because the heat generation inside the composite and the heat loss to the surroundings reached a balance. The drop at the beginning of the dwell stage was ascribed to the heat conduction from the hot spots to the cold spots inside the composite plate. During the random field variation heating, the tendency of the maximum temperature difference is similar as that of the single pattern heating, but the ramp rate is much smaller (especially in the marked section) due to the randomly complementary effect of extensive HPs. As a consequence, the temperature distribution during the random field variation heating is more homogeneous than that in the single pattern heating. Unlike the first two heating processes, the results of the multi-pattern compensation heating are totally different. As shown, when the maximum temperature difference exceeded 6°C (preset threshold) the HP compensation diagram was triggered. Hence, the curve fluctuated around 6°C for some time. Then, there was a small overshoot along with the rise of the composite temperature, but it was gradually adjusted back by the system. Simultaneously, it can be found that the continuous compensation process also makes the curve much more rough than the other two heating process. After that, the curve overshoot several times but was adjusted back one by one until the end of the processing. This is because this paper only considers the hottest spot or the coldest spot during one compensation step, which is difficult to keep the maximum temperature difference stabilizing at 6°C at relatively high temperature levels. Even then, the temperature difference was reduced considerably, which effectively validates the feasibility of the proposed multi-pattern compensation method. Further reductions in the temperature difference of composite parts can be realized by considering the hottest spot and the coldest spot, even the secondary hot spot and the secondary cold spot et al, simultaneously during one compensation step.

6. Conclusions

Based on the authors' experimental findings that there is a one-to-one correspondence between composite HPs and MCSSs, a multi-pattern compensation method was proposed to realize a homogeneous microwave curing process for advanced

composite materials. In order to ensure the feasibility of the multi-pattern compensation method, the theoretical analysis of the findings was investigated; the principle of this method was discussed; and the control strategy of this method is designed. It was demonstrated to be a feasible plan through the comparison of resulting temperature difference between this and other two traditional microwave curing process. Under the situation of only considering the hottest spot or the coldest spot during one compensation step, the maximum temperature difference of a short carbon fiber/epoxy composite plate was reduced by 67% and 58% compared with the traditional single pattern heating and random field variation heating process. Further reductions in the temperature difference of composite parts can be realized by considering the hottest spot and the coldest spot, even the secondary hot spot and the secondary cold spot et al, simultaneously during one compensation step. This technology can be potentially used in other microwave heating processes as well where a high temperature uniformity is required.

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References

- [1] Zhang X, Fan X, Yan C, Li H, Zhu Y, Li X, Yu L. Interfacial microstructure and properties of carbon fiber composites modified with graphene oxide. *ACS applied materials & interfaces* 2012; 4(3): 1543-1552.
- [2] Gaurav A, Singh K K. Fatigue behavior of FRP composites and CNT-Embedded FRP composites: A review. *Polymer Composites* 2016. DOI: 10.1002/pc.24177.
- [3] Mallon P J, O'Bradaigh C M. Development of a pilot autoclave for polymeric diaphragm forming of continuous fibre-reinforced thermoplastics. *Composites* 1988;

- 19(1): 37-47.
- [4] Yanagimoto J, Ikeuchi K. Sheet Forming Process of Carbon Fiber Reinforced Plastics for Lightweight Parts. CIRP Annals-Manufacturing Technology 2012; 61/1:247-250.
 - [5] Joshi S C, Bhudolia S K. Microwave–thermal technique for energy and time efficient curing of carbon fiber reinforced polymer prepreg composites. Journal of Composite Materials 2014; 48/24:3035-3048.
 - [6] <http://aviationweek.com/paris-air-show-2015/airbus-plans-increase-a350-production-rate-13-month>
 - [7] Zhou J, Li Y, Li N, Hao X. Enhanced interlaminar fracture toughness of carbon fiber/bismaleimide composites via microwave curing. Journal of Composite Materials 2017; 51(18): 2585-2595.
 - [8] http://www.terruzzifercalxgroup.com/en/prod_autocl_composite.php
 - [9] Johnston A A. An integrated model of the development of process-induced deformation in autoclave processing of composite structures. University of British Columbia, 1997.
 - [10] Papargyris D A, Day R J, Nesbitt A, Bakavos D. Comparison of the mechanical and physical properties of a carbon fibre epoxy composite manufactured by resin transfer moulding using conventional and microwave heating. Composites Science and Technology 2008; 68(7): 1854-1861.
 - [11] Li N, Li Y, Hao X, Gao J. A comparative experiment for the analysis of microwave and thermal process induced strains of carbon fiber/bismaleimide composite materials. Composites Science and Technology 2015; 106:15-19.
 - [12] Thostenson E T, Chou T W. Microwave processing: fundamentals and applications. Composites Part A: Applied Science and Manufacturing 1999; 30(9):1055-1071.
 - [13] Johnston K, Pavuluri S K, Leonard M T, Desmulliez M P Y, Arrighi V. Microwave and thermal curing of an epoxy resin for microelectronic applications. Thermochimica Acta 2015; 616:100-109.

- [14]Zhou J, Li Y, Li N, Hao X, Liu C. Interfacial shear strength of microwave processed carbon fiber/epoxy composites characterized by an improved fiber-bundle pull-out test. *Composites Science and Technology* 2016; 133:173-183.
- [15]Thostenson E T, Chou T W. Microwave and conventional curing of thick-section thermoset composite laminates: Experiment and simulation. *Polymer composites* 2001; 22(2): 197-212.
- [16]Nightingale C, Day R J. Flexural and interlaminar shear strength properties of carbon fibre/epoxy composites cured thermally and with microwave radiation. *Composites Part A: Applied Science and Manufacturing* 2002; 33(7): 1021-1030.
- [17]Li N, Li Y, Zhou J, He Y, Hao X. Drilling delamination and thermal damage of carbon nanotube/carbon fiber reinforced epoxy composites processed by microwave curing. *International Journal of Machine Tools & Manufacture* 2015; 97:11-17.
- [18]Kwak M, Robinson P, Bismarck A, Wise R. Microwave curing of carbon-epoxy composites: Penetration depth and material characterisation. *Composites Part A: Applied Science and Manufacturing* 2015; 75: 18-27.
- [19]Mishra R R, Sharma A K. Microwave-material interaction phenomena: Heating mechanisms, challenges and opportunities in material processing. *Composites Part A: Applied Science and Manufacturing* 2016; 81: 78-97.
- [20]Sturm G S J, Verweij M D, Van Gerven T, Stankiewicz A I, Stefanidis G D. On the parametric sensitivity of heat generation by resonant microwave fields in process fluids. *International Journal of Heat and Mass Transfer* 2013; 57(1): 375-388.
- [21]Sturm G S J, Verweij M D, Van Gerven T, Stankiewicz A I, Stefanidis G D. On the effect of resonant microwave fields on temperature distribution in time and space. *International Journal of Heat and Mass Transfer* 2012; 55(13):3800-3811.
- [22]Kimrey H D, Janney M A. Design principles for high frequency microwave cavities. In: Sutton W H, Brooks M H, Chabinsky I J, editors. *Microwave processing of materials*, Materials Research Society Proceedings, 124. Pittsburgh: Materials Research Society 1988; 367-372.

- [23] Tran V N. An applicator design for processing large quantities of dielectric materials. In: Clark D E, Gac F D, Sutton W H, editors. *Microwaves: theory and application in materials processing*, ceramic transactions, Proceedings of the symposium held during the 93rd Annual Meeting of the American Ceramic Society, Cincinnati, 21.
- [24] Risman P O, Ohlsson T, Wass B. Principles and models of power density distribution in microwave oven loads. *Journal of Microwave Power and Electromagnetic Energy* 1987; 22(4): 193-198.
- [25] Qiu Y, Hawley M. Uniform processing of V-shaped and tri-planar composite parts using microwaves. *Journal of composite materials* 2001; 35(12): 1062-1078.
- [26] Demuse M T, Johnson A C. Variable frequency microwave processing of thermoset polymer matrix composites. In: Iskander M F, Lauf R J, Sutton W H, editors. *Microwave processing of materials IV*, Materials Research Society Proceedings, San Francisco, 347. Pittsburgh: Materials Research Society 1994; 723-727.
- [27] Li Y, Hang X, Li N, Hao X. A temperature distribution prediction model of carbon fiber reinforced composites during microwave cure. *Journal of Materials Processing Technology* 2016; 230:280-287.
- [28] Zhou J, Li Y, Cheng L, Hao X. Dielectric properties of continuous fiber reinforced polymer composites: modeling, validation and application. *Polymer Composites* 2017; DOI: 10.1002/pc.24579.
- [29] Wang Z J, Yin Z, Xiong Y L. Temperature control and PID parameters optimization based on finite element model. *IEEE* 2010:2241-2244.

Figure Captions

Fig.1. Illustration of one-to-one correspondence between composite HPs and MCSSs.

Fig.2. Idea of the multi-pattern compensation method.

Fig.3. Control system of the multi-pattern compensation method.

Fig.4. Illustration for composite HP database construction.

Fig.5. Strategy for composite HP collection.

Fig.6. Demonstration for the effectiveness of the microwave power allocation strategy.

Fig.7. Photograph of (a) the octagon microwave curing equipment, (b) infrared thermal imager and (c) short carbon fiber/epoxy composite plate.

Fig.8. Schematic illustration of (a) rectangular microwave oven, (b) electric field distribution of TE_{101} and TE_{102} modes, (c) resonant applicator before shape perturbation, (d) resonant applicator after shape perturbation, (e) resonant applicator before material perturbation and (f) resonant applicator after material perturbation.

Fig.9. Temperature profiles of (a) traditional single pattern heating, (b) random field variation heating and (c) multi-pattern compensation heating.

Fig.10. Heating preference of the composite plate at different temperatures during traditional single pattern heating.

Fig.11. Heating preference of the composite plate at different temperatures during random field variation heating.

Fig.12. Heating preference of the composite plate at different temperatures during multi-pattern compensation heating.

Fig.13. Statistics of the maximum temperature difference of the traditional single pattern heating, random field variation heating and multi-pattern compensation heating.