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INVESTIGATION OF DIRECTIONAL THERMAL DIFFUSIVITY FOR GRAPHITE COMPOSITE

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Abstract

Orthotropic thermal diffusivity (TD) of an aircraft construction/repair composite is discussed. The material concerned is an epoxy resin matrix with graphite fabric filling supplied as a prepreg sheet. The out-of-plane (cross-plane) and in-plane TD has been measured for about 1.4 mm thick flat specimens by applying modified Ångström's method. In the case of longitudinal measurements, two characteristic directions have been concerned: parallel to fabric fibre and bisectional to the fabric interlace. The thermal diffusivity has been determined in function of the temperature within the temperature interval from 0℃ to 70℃. Due to a difference in characteristic dimensions while performing longitudinal and out of plane measurements, two different approaches in application of the Ångström's method have been used. Because of the investigated surface roughness, the obtained results should be treated as the measured i.e. apparent ones. The study has shown a significant difference in out-of-plane and in plane measured TD values. On the contrary, the fibre parallel and bisectional TD values have been revealed almost the same.

Keywords: *orthotropic thermal diffusivity, graphite composite, temperature oscillation method*

1. Introduction

There are many drivers for development of materials for airframes. One of them is a weight saving through increased specific strength or stiffness. The fibre composite approach can provide significant improvement in those parameters over metal alloys [1, 2]. While ensuring similarity in many properties the composite structure can differ in others. A substantial difference is typically observed in thermal transport properties characterized by the thermal conductivity or the thermal diffusivity (TD). The difference concerns both the quantity as well as quality. In particular, fibre, reinforced composites usually exhibit significant directional differences – anisotropy – that is not so common in metallic materials. These differences need a proper characterisation. However, regarding that composites are mostly applied in shell structures the characteristic transversal and longitudinal dimensions of a certain investigated material sample can vary for even two ranges of magnitude. This impedes experimental studies, especially measurements of thermophysical property (TP) as thermal diffusivity or thermal conductivity. For that, reason dedicated methods and measurement procedures should be applied to get reliable and fully compatible result.

The present article is devoted do discussion of the directional thermal diffusivity measurements that were performed for a certain fabric composite material prepared from a prepreg. The investigated material is applied for the airframe component fabrication and repair. Because of the

above-mentioned difficulties, the modified Ångström's method was applied for the thermal diffusivity measurements [3-8]. The method takes advantage from the temperature oscillation and can be accommodated to both transversal (out-of-plane) and longitudinal (in-plane) thermal diffusivity measurement. The TD studies were complemented with gravimetric and microcalorietric measurements that enabled for the thermal conductivity calculation.

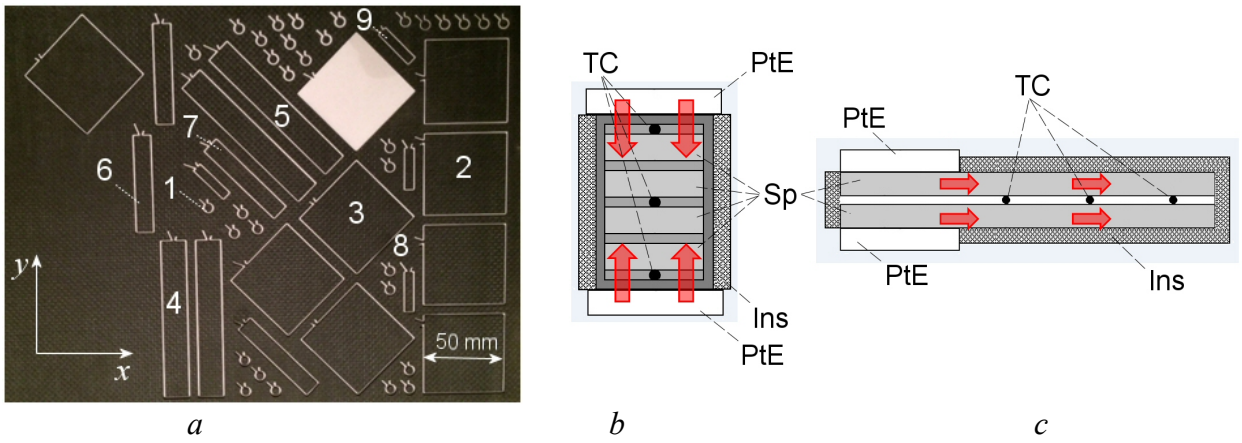
2. Experimental

Specimens of the epoxy resin matrix with graphite fabric filling were prepared from a 1.38 mm thick plate made from a prepreg sheet. The specific prepreg was a BMS8-168 type material comprising 4 unidirectionally layered plain woven 250F graphite fabric layers (at CL875 configuration). The prepreg and the epoxy resin manufacturer was a Hexcel company while graphite fibres were manufactured by a Cytec firm. The material for investigation was cured for 150 min. at 121°C. The heating and cooling rate for the isothermal heat treatment step was equal to ±3 K/min. Specimens for thermophysical property investigations were cut by applying a water jet. A proper orientation was preserved as illustrated in Fig. 1a. Specimens for the cross-plane TD measurements were prepared as plates of 50 mm sides (Fig. 1a: No 2, 3), while specimens for the in-plane TD measurements were produced as rectangular of 15 mm and 100 mm sides. The appropriate parallel and bisectional orientations are denoted as 0° (Fig. 1a: No 4) and 45° orientation (Fig. 1a: No 5). Microcalorimetric DSC studies were performed applying disc shape specimens of 5 mm diameter (Fig. 1a: No 1).

Nevertheless that the present study is focussed on TD investigations the experiments started from gravimetric and microcalorimetric measurements. It was not only because of a need for a proper material characterization but also for testing the material property thermal stability while performing DSC thermal analyses [9].

The thermal analysis was conducted using a PerkinElmer Pyris 1 DSC. Investigations were performed within a range from -20°C to 130°C on thermal cycling that means that heating and cooling cycles were repeated at least once. A dynamic dehydrated N2 atmosphere with a flow rate of 20 ml/min. was employed. Low temperature measurements were possible due to the use of Intracooler 1 attachment. During investigations the attention was focused mostly on specific heat i.e. c_p temperature dependence. The ratio method ("three curve method") [10] with dedicated procedure of divided heating/curing steps [11] was utilised. The temperature rate was equal to ±5 K/min., the isothermal 1 min. stops were at -20°C, 20°C, 90°C, 120°C and 130°C. A sapphire reference specimen of 60.33 mg mass was applied as a reference.

Periodic heating or so-called temperature (thermal) wave technique is probably the oldest one transient technique for investigation of thermophysical properties. Initially described by Ångström in 1861 in application for investigation of metal bars [3] it was being many times modified and accommodated for measurements for specimens of different shapes and types. At this particular instance the procedure and apparatus described in [7] and [8] was utilised. Configuration of the investigated specimens for cross plane and two directions of in-plane TD investigations are shown in Figs 1b and 1c respectively. When performing measurements the space between Peltier elements and the investigated specimen surfaces was filled with a thermally conductive DowCorning paste for a better thermal contact. All studies were done on thermal cycling within the temperature range from -20°C to 70°C while measuring flat specimens (Fig. 1a, No 2) for transversal TD and from -10°C to 80°C for longitudinal TD on slab specimens cut at 0° and 45° directions (Fig. 1a, No 4 and 5). In both cases, duplicated specimens were applied (Fig. 1b and c). The heating/cooling rates for these measurements were equal to ±0.4 K/min. and ±0.25 K/min. Type K (Omega) thermocouples of 0.05 mm wires were utilised for the temperature evolution measurements. The thermal diffusivity was from both the amplitude attenuation and phase shift of the response signals (see e.g. [8] giving independently simultaneous values of so called the amplitude a_{ampl} and the phase a_{phase} TD in result of the temperature recordings analyses.



a *b* *c*

Fig. 1. Illustration of the graphite composite specimens preparation (a) and scheme of TD specimens configuration for out-of-plane (b) and in-plane (c) TD components studies. The numbers and symbols indicate: 1 – DSC specimen, 2, 3 – TD specimen for out-of-plane study, 4 – specimen for in-plane TD study in parallel to fibres direction (0° direction), 5 – specimen for in-plane TD study bisectionally (45° direction), 7, 8, 9 – specimens for DMA (Dynamic thermo-Mechanical Analysis) and Dil (dilatimetric) measurements, Sp – investigated specimen, TC – thermocouples, PtE – Peltier elements, Ins – insulation. The arrows indicate direction of the temperature oscillation evolution

3. Results and discussions

Prior to TP investigations, a representative value of a density of the cured composite was established in course of gravimetric measurements. The task was done applying a Mettler-Toledo AT 261 microbalance equipped with a density kit. A distilled water was applied as immersing fluid. Measurements were performed on TD, DMA (Fig. 1a: No. 6, 7) and Dil (Fig. 1a: No. 8, 9) specimens[1]. The mass weighted average from these measurements is shown in Tab. 1.

As it commonly known the prepreg composites usually, exhibit roughness of upper and bottom surfaces. In order to characterize this roughness, in addition to density measurements, calculations of the apparent density were performed. They were based on the mass measurements done with the same microbalance and on the contour volume evaluation from linear dimensions measurements performed for specimens of types 2-9 (Fig. 1a). The contour thicknesses were measured with a micrometre screw. The two obtained density values differ for 9.2%.

Tab. 1. Comparison between the gravimetrically measured density and the apparent density calculated from contour volume of the investigated specimens with rough upper and bottom surfaces

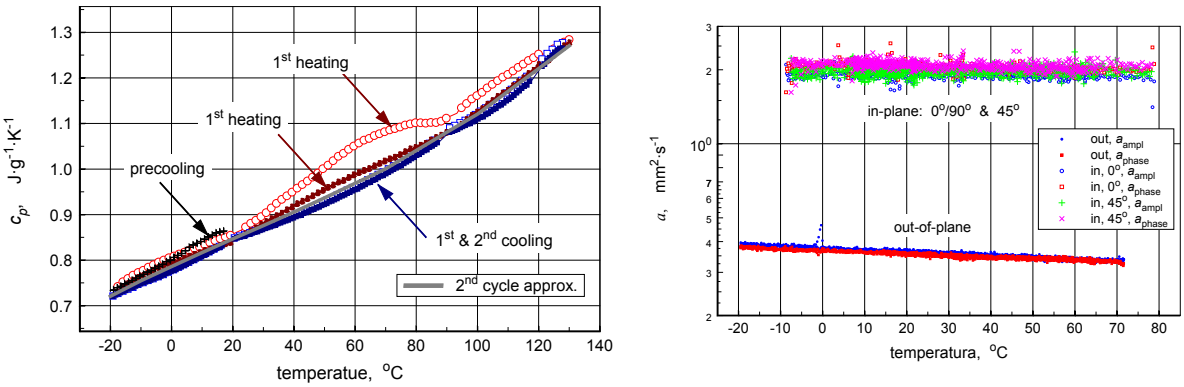
Density ρ(20°C), kg·m^{-3}	Apparent density ρ_{app}(20°C), kg·m^{-3}
1468 ± 31	1333 ± 62

The results of microcalorimetric studies are depicted in Fig. 2a. The analysis of the obtained data reveals irreversibility in the 1st heating. The access in specific heat value could be attributed to the heat consumption due to the absorbed moisture release. The post-curing effects are less probable because of the discrepancy in specific heat data substantially decreases above 80°C, far below the curing temperature equal to 121°C. The obtained c_p values suggest high graphite fibres contents in the investigated composite – they are more close to the carbon specific heat then to the epoxy resin type material specific heat (comp. e.g. [12] and [13]). By applying the raw specific heat, data from 2nd heating and cooling a representative characteristic was derived by least square approximation. A 2nd polynomial fit was applied and the appropriate results are shown in Tab. 1.

Figure 2b illustrates results of the TD studies. The data has not been corrected for the thermal

[1] *The specific DMA and Dil measurements are not discussed here.*

expansion. For all three types of the investigated specimens, both the amplitude and phase TD values were obtained. The a_{ampl} and a_{phase} should be the same when the model heat transfer conditions are preserved. In the case of any departures from model conditions, the amplitude and phase values deviate [2, 6, 8]. However, the real TD value is bounded by the obtained two experimental estimates, so the difference could be used as a measure of any systematic error. I our case obtained amplitude and phase TD values overlap each other within the relative difference smaller than 5%.



a *b*

Fig. 2. Results of DSC measurements of specific heat (a) and results of the directional TD studies by applying temperature oscillation (b)

The results showed a distinct difference in transversal and longitudinal TD. The out-off-plane TD was observed to be about one fifth of the in-plane TD. According to the obtained values they agree with results of investigations reported in [14] for similar types of composites. The TD measured in 0° and 45° directions revealed to be, as had been expected, the same within the measurement uncertainty limits. Nevertheless, just for illustration purposes, the approximates from the thermal diffusivity measurements were calculated for every studied case separately (Tab. 2). The geometrically averaged amplitude and phase data were fitted with linear functions. Tabulated data from approximations are displayed in Tab. 2. It should be mentioned that the TD data were extrapolated to complement DSC results – these figures are indicated by italic typing.

On the basis of the obtained results, the representative directional thermal conductivity values were calculated according to the following formulae:

$$\lambda(t) = \rho_0 \, a(t) \, c_p(t), \tag{1}$$

where λ is the thermal conductivity, t – temperature and $\rho_0 = \rho(20°C)$ – the room temperature density (comp. Tab. 1). As one can notice the apparent thermal conductivity was not been corrected for the thermal expansion also. However, in this particular case, the in-plane TD data have treated in total and the only one transversal thermal conductivity value was obtained for every single temperature point.

It should be underlined once again that the obtained TD and the thermal conductivity data should be treated as apparent ones. It is because of roughness of the investigated specimens. The roughness could not be eliminated by polishing, as this process will destroy the composite structure. However rough estimates made on the basis of density data shown in Tab. 1 prove that the corrections should not exceed 5% for in-plane TD and 10% for out-of-plane TD. The measured transversal TD is overestimated while the longitudinal TD is underestimated. The same concerns thermal conductivity calculation results. Nevertheless, the obtained data are of the primary importance regarding the aviation technology needs.

Tab. 2. Results of thermophysical property measurements (specific heat, thermal diffusivity – approximated data) and calculation (apparent thermal conductivity – by applying Eq. for the density value $\rho(20℃)=1468\ kg \cdot m^{-3}$)

Temperature t	Specific heat	Thermal diffusivity			Thermal conductivity	
t, °C	c_p, $J \cdot g^{-1} \cdot K^{-1}$	a, $mm^2 \cdot s^{-1}$			λ, $W \cdot m^{-1} \cdot K^{-1}$	
		out-of-plane	in-plane		out-of-plane	in-plane
			0°	45°		
-20	0.720	0.384	*2.025*	*2.051*	0.406	*2.15*
-10	0.754	0.379	2.014	2.047	0.419	2.25
0	0.785	0.373	2.004	2.044	0.430	2.33
10	0.816	0.367	1.993	2.040	0.439	2.41
20	0.846	0.361	1.982	2.036	0.448	2.49
30	0.875	0.355	1.972	2.032	0.456	2.57
40	0.905	0.349	1.961	2.028	0.464	2.65
50	0.936	0.344	1.950	2.024	0.472	2.73
60	0.968	0.338	1.940	2.020	0.480	2.81
70	1.002	0.332	1.929	2.016	0.488	2.90
80	1.038	*0.326*	1.919	2.012	*0.497*	3.00
90	1.077	*0.320*	*1.908*	2.008	*0.506*	*3.10*
100	1.120	*0.314*	*1.897*	2.004	*0.517*	*3.21*
110	1.167	*0.308*	*1.887*	2.000	*0.528*	*3.33*
120	1.218	*0.303*	*1.876*	1.996	*0.541*	*3.46*

3. Conclusions

Application of dedicated DSC and modified Ångström's method measurement procedures enabled for obtaining complemented characteristics of thermal properties of the epoxy resin matrix and graphite plain-woven fabric filling composite. The investigations were focused on directional differences in thermal transport properties of the studied material. On the basis of the measured density, specific heat and in-plane and out-of-plane thermal diffusivity a temperature dependence of the thermal conductivity was calculated for every characteristic direction. It was shown that the longitudinal thermal conductivity is for about 5-6 times greater than transversal thermal conductivity. However, the revealed directional difference of transversal and longitudinal thermal transport parameters for the composite are not so distinct as those observed for anisotropic graphite structures [1, 2], in that range graphite fibres used in the composite plain woven fabric filling.

Because the discussed results are obtained just at first stage of a wider research, they should be analysed more thoroughly for the revealed phenomena. Such an analysis needs to be based on additional experimental data, in that range on the results of dilatometric measurements. The appropriate investigation programme is in progress and the results will be reported soon.

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