

PROCESSING OF LOW-VISCOSITY CBT THERMOPLASTIC COMPOSITES: HEAT TRANSFER ANALYSIS

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ABSTRACT: Cyclic butylene teraphthalate (CBT) thermoplastics offer significant advantages in terms of composite processing. Primarily, their inherent low viscosity means that fibre wet-out and impregnation can occur easily and rapidly, offering potential reductions in processing time and cost. CBT composites can offer the benefits of thermoplastics, including toughness and recyclability, without the high viscosities normally associated with these materials. These advantages are particularly significant for the manufacture of large-scale composite structures, for example, wind turbine blades. Existing technology using thermosets mean that it is getting more and more difficult to process large quantities of composite materials cost-effectively.

However, there are certain issues peculiar to CBT thermoplastics that must be overcome before large-scale processing can occur. High temperatures and rapid heating rates are unique to these materials when compared to traditional thermosets. The nature of the heat transfer mechanism for CBT materials heated this way is not very well understood. Previous experimental work in this area has provided some degree of understanding. The effect of different forms of material (pre-preg and power-impregnated dry glass fibres) has been investigated.

The heating mechanism of the various material forms is discussed and analysed. The temperature-dependent thermal conductivity of the various material forms is quantified using a guarded comparative longitudinal heat flow apparatus. A one-dimensional transient heat transfer model is obtained which will assist in determining if the necessary processing conditions are met before CBT processing is carried out.

KEYWORDS: Cyclic butylene teraphthalate (CBT) composites, resin film infusion, powder impregnation, composite tooling, heating mechanism, thermal conductivity.

INTRODUCTION

CBTTM (cyclic butylene teraphthalate) is a relatively new material that has been developed by Cyclics Corp. CBT is based upon using the cyclic oligomer form of PBT, a conventional engineering thermoplastic. PBT's natural properties mean that it has very good potential to be used in a range of thermoplastic composite applications [1]. Processing of CBT occurs when a catalyst is added to the material and when subsequently heated, the short-chain oligomer chains open and bond together to form the long-chain polymeric PBT-form of the material. The inherent low initial viscosity (circa. 150 mPa.s), resulting from the use of shorter chain oligomers, means that it is eminently suitable for rapid processing of thermoplastic composites. Previous applications for thermoplastic composites have been restricted by having to use much higher temperatures and pressures in order to 'wet-out' reinforcing fibres using the much higher viscosity polymer-form of the thermoplastic.

CBT-composites may be processed in a number of ways; however, two methods are most pertinent to the work that is discussed in this paper: resin film infusion / pre-preg and powder impregnation [2] (Figure 1). Resin film infusion (RFI) uses continuous carbon or glass fibre reinforcement that has been 'pre-impregnated' with the CBT-oligomer form of the material prior to being processed. A special catalyst is added to the CBT material that is not activated until a set temperature is reached. In essence, the CBT forms a resin 'film' mainly on one side of the reinforcement fabric that only fully 'wets-out' the fibres when secondary processing occurs at pressure. A more appropriate term to use would be 'semi-preg' rather than 'pre-preg' as only one side of the glass fabric is completely coated with CBT during the application stage. Plies may be stacked in a conventional manner and 'de-bulk' significantly when heat and pressure are applied during processing.

Powder-impregnated processing of CBT involves using dry continuous fibre reinforcements that are 'sandwiched' together, with an intervening layer of CBT dry powder between reinforcement layers. The powder is laid down during the lay-up process. Although more difficult to handle, powder impregnation does offer potential in terms of cost reduction (no need for additional pre-preg creation stage) and allows easier manipulation of the dry fabric layers as they are much more flexible compared to the 'boardy' and rigid RFI material. The higher bulk density of the powder also means that there is a more significant amount of de-bulking and thickness reduction that occurs for powder-impregnated CBT.

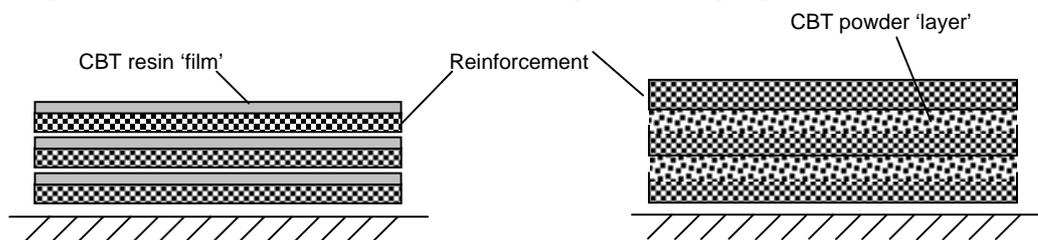


Figure 1 Simple schematic of RFI and powder-impregnated CBT/glass fibre forms

A heat transfer analysis will be especially important going forward for CBT processing in order to design appropriate heated tooling and to allow prediction of the temperature profiles, heat transfer rates, etc. throughout the thickness of CBT-composites. This will be essential for very large thermoplastic composites structures e.g. wind turbine blades, where thick root section thicknesses (greater than 100mm) and use of sandwich construction in the aerofoil sections greatly complicates the heat transfer issue [3].

HEAT-UP EXPERIMENTAL ANALYSIS

It is useful initially to carry out a direct comparison of 3 different material forms as regards heat-up characteristics during processing – dry glass fibres (reference), RFI and powder-impregnated glass fabric. To do this, a series of simple heat-up experiments was carried out. This involved placing a typical 6-layer lay-up on the surface of a heatable metal platen within a vacuum bag, attaching thermocouples at different points throughout the thickness and applying heat to a typical set-point of 195°C for CBT. The initial thickness of the dry glass fibre lay-up (using Saertex UD non-crimp glass fabric, 950 gsm) when vacuum is applied was 4.5 mm. Results for the dry glass only are shown in Figure 2. As expected, the temperatures within the glass fibres lag the platen temperature by up to 20°C. However, the application of vacuum pressure ensures that intimate contact is made between the glass fibre layers and heat transfer occurs quite efficiently by conduction.

Figure 3 shows the heat-up curve for a CBT-resin film infused glass fabric lay-up using the same type of glass reinforcement in terms of reinforcement type (unidirectional E-glass, 950 gsm). The initial stack thickness is significantly greater than with the dry glass, approximately 7.5 mm. This time, there is a much more significant temperature lag initially. The RFI layers are boardy and inflexible, even under vacuum pressure. Thermal contact resistance is much higher compared to dry glass due to the semi-rigid nature of the RFI form and non-intimate contact between the layers. Up to 120°C, the temperature of the top layer lags the bottom layer by up to 40°C. However, as the temperature of the stack approaches and passes 150°C, the heat transfer mechanism is significantly altered by a change in state of the CBT. It begins to melt prior to polymerisation taking place at higher temperatures. This phase change increases the heat transfer rate suddenly as the layers come into much more intimate contact as the liquid form of CBT removes any gaps between the layers and de-bulking occurs causing the thickness of the stack to reduce significantly. This sudden increase in heat transfer is seen as a ‘kink’ upwards on the time-temperature curve at around 130°C. The ‘kink’ is somewhat reversed between 140°C and 150°C (decrease in heat transfer rate) due to the latent heat of fusion effect as all of the CBT melts. Above 160°C, the through-thickness temperatures converge rapidly until the difference is only approximately 5-6°C at 190°C.

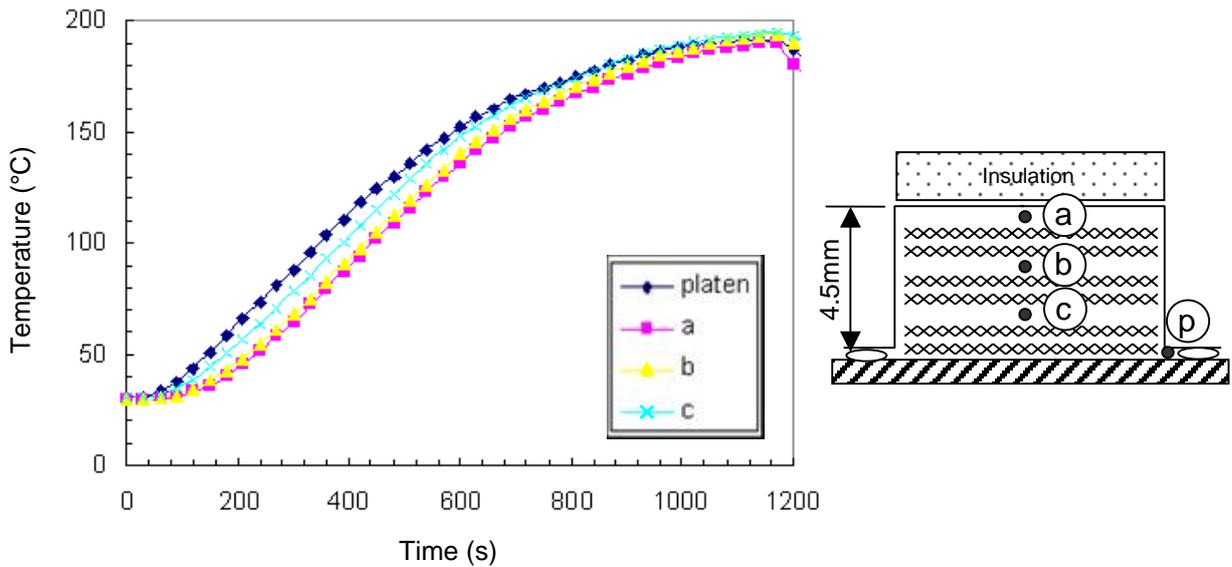


Figure 2 Heat-up of dry glass fibres

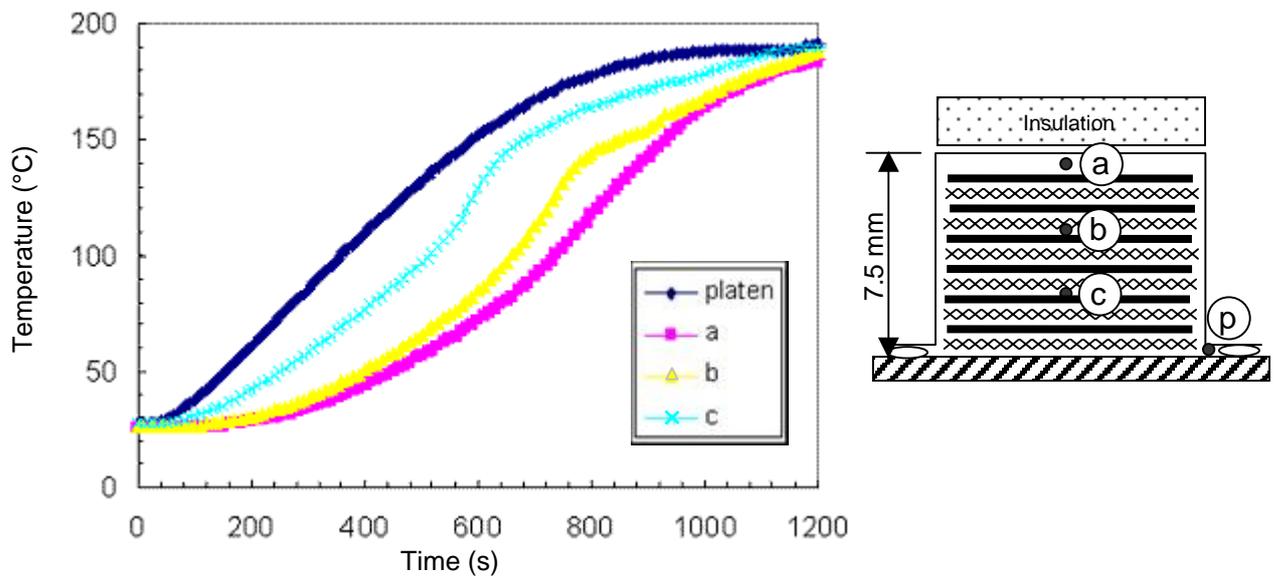


Figure 3 Heat-up of RFI/CBT-glass fibre layers

For the powder-impregnated form, the glass fibre layers (unidirectional E-glass, 950 gsm) were placed on the platen, with the CBT powder ‘sandwiched’ in between. The amount of CBT powder applied was the same amount as that used for the RFI-form in order to produce a final glass fibre volume fraction of 50%. Five identical powder layers were applied between the six glass fibre layers. When vacuum pressure was applied, the much greater bulk density of the CBT-powder meant the initial stack thickness was 10.4 mm, a 35% increase in thickness. It might therefore be expected that the powder impregnated form would exhibit the poorest heat transfer performance due to the greater thickness. The time–temperature heat-up curve is shown in Figure 4. Again, the through-thickness temperatures lag the platen temperature significantly. However, compared to Figure 3 (RFI-form), the heat transfer performance is significantly improved. 100°C is reached at Thermocouple A (top of the stack) after approximately 500s (8.3°C/min) compared to 720s for the RFI-form (5.8°C/min). This

may be attributed to a much greater intimate contact between the layers for the powder-impregnated form, allowing heat transfer by conduction to occur more readily, despite the increased thickness. Again, the latent heat of fusion effect as the CBT melts is seen as a kink 'downwards' in the slope of the graph at about 140-150°C. After this however, the temperatures converge rapidly as heat transfer occurs more readily through the fully 'wetted-out' glass fibre layers.

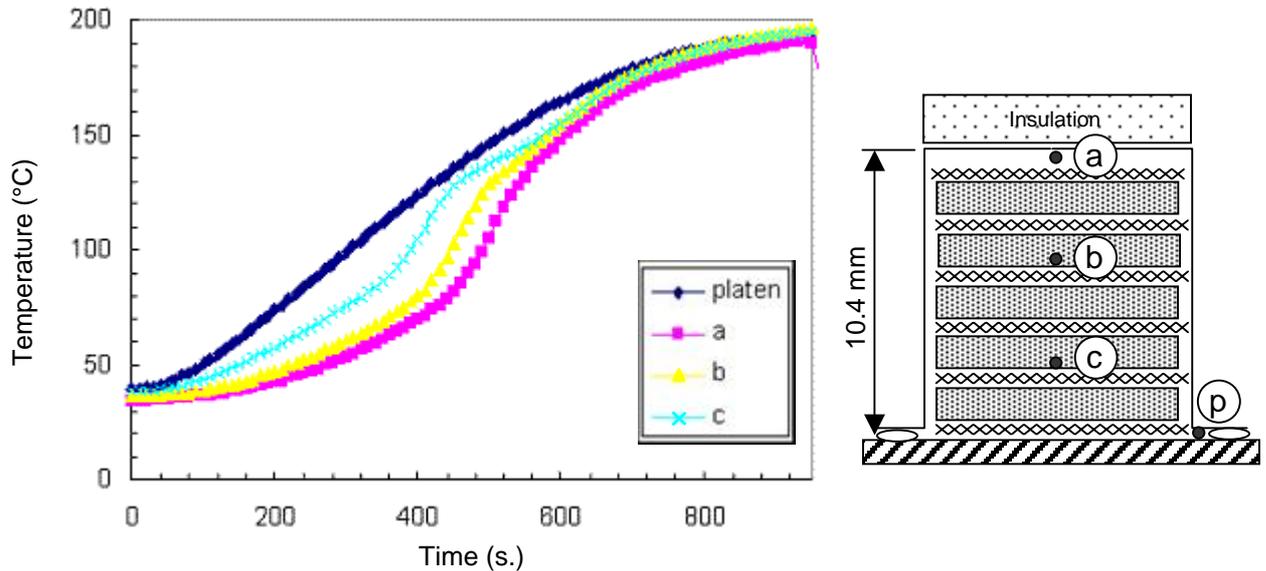


Figure 4 Heat-up of powdered CBT/glass fibre layers

THERMAL CONDUCTIVITY MEASUREMENT

The behaviour of CBT composites is quite complex as regards heating behaviour. It has been demonstrated how the presence of CBT (as either a resin film or powder) can initially insulate and prevent heat flow, then as the CBT begins to melt, heat flow is increased significantly. A detailed analytical study incorporating a non-linear model of phase change effects on the heat transfer is beyond the scope of this work. Instead, an alternative approach is taken in which an account is made of temperature-variant thermal conductivity with the lay-up during heating – this is a reasonable assumption to make if we assume that the lay-up stack including all of the glass fibre and CBT behaves as a solid continuum during heating.

In order to measure thermal conductivity, an apparatus designed and built by Lee [4] was used. This apparatus uses the guarded comparative longitudinal heat flow technique to measure 'k' values for different materials compared to a reference block of known conductivity. The design is based on a standard test method (ASTM E 1225-87). Although Lee only used his apparatus for measuring the thermal conductivity of dry fibre stacks, it was considered feasible to use it also for CBT RFI 'pre-preg' and CBT-powder impregnated glass fibre.

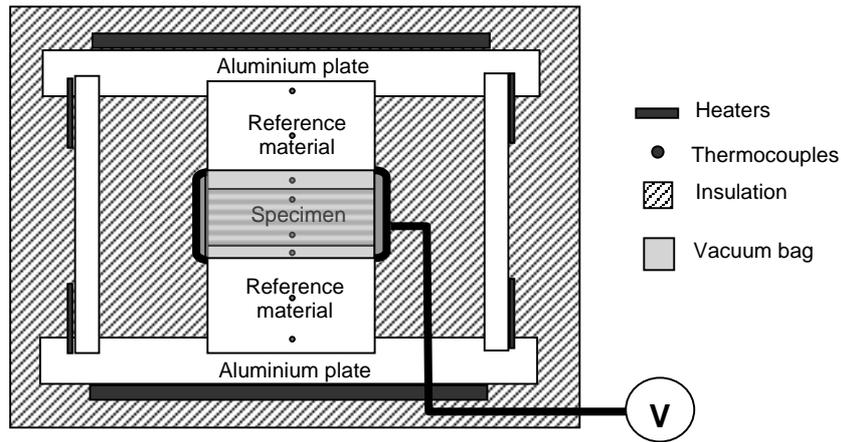


Figure 5 Schematic of thermal conductivity measurement apparatus

A schematic of the thermal conductivity test apparatus is shown in Figure 5. The test specimen (in our case the CBT/glass fibre combinations) was placed between the reference blocks. In order to minimise contact resistance between the top and bottom layers and the reference material surfaces, a layer of thermally conductive paste was applied. A vacuum bag was applied circumferentially around the stack. When vacuum was applied, this had the effect of compressing the stack, similar to that seen on the heated platen from the previous set of experiments. A set-point temperature was specified for both the top and bottom heaters and a steady-state temperature gradient was allowed to develop over a period of time. The ‘k’ value for the specimen material was calculated by comparing to a known conductivity for the Macor glass ceramic reference blocks at a particular temperature.

Results are shown in Figure 6. The ‘k’ values for the dry glass fibres (GF) were taken from [4]. The thermal conductivity for the RFI prepreg and the powder impregnated glass was measured at 90°C and 120°C. Due to wattage restraints on the equipment, it was not possible to measure thermal conductivity at higher temperatures. This is unfortunate as it is above 120°C that the thermal properties of CBT begin to vary significantly as it changes from a solid to a liquid.

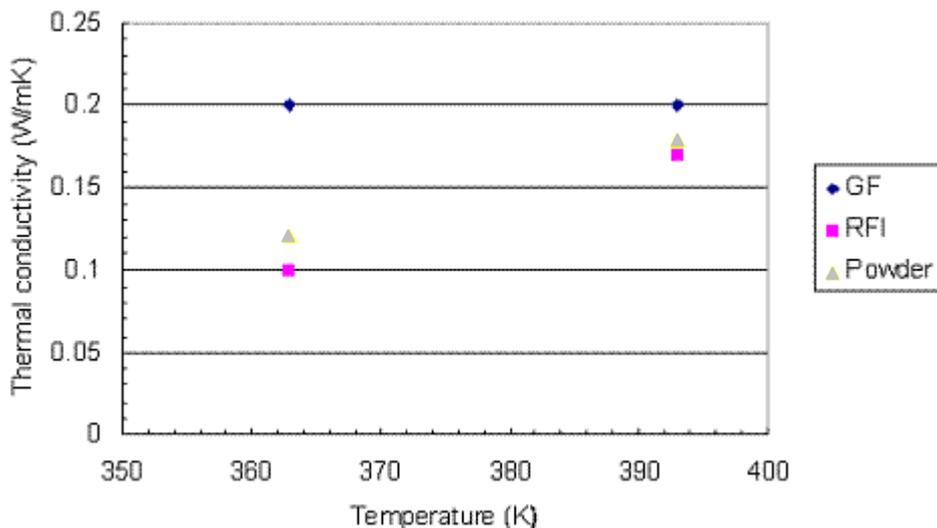


Figure 6 Thermal conductivity for material forms

1-D TRANSIENT HEAT TRANSFER MODEL

In general terms, the heating of a flat laminate with initial temperature T , thickness x , much smaller than its length and width, can be described as a simple 1-D heat conduction problem. Certain assumptions need to be made such as constant temperature boundary conditions, homogeneous make-up of the temperature slab, initially uniform slab temperature, etc. For our purposes, it will be useful to have a model which correlates experimental data with the predictive model using thermal conductivity data obtained in the previous section.

The standard law of thermal conservation of energy applies and the heat transfer equation may be written as [5]:

$$\frac{\partial}{\partial t}(\rho c_p T) = \frac{\partial}{\partial x} \left(k \frac{\partial T}{\partial x} \right) - Q \quad (1)$$

where ρ = material density, c_p is the specific heat, k is the thermal conductivity and Q represents the heat loss due to the latent heat of fusion upon melting.

It is assumed for the initial model that k is a function of temperature and that ρ and c_p are relatively constant with temperature. Also, we will only be modelling the heat transfer up to 120°C, prior to the phase change of the material, therefore Q can be considered to be zero. A more comprehensive model will be developed as part of future work to incorporate heating to higher temperatures.

Eqn. (1) is a partial differential equation and may be solved using the finite difference technique:

$$\frac{\partial T}{\Delta t} = \frac{k}{\rho c_p} \left(\frac{\Delta(\Delta T)}{\Delta x^2} \right) \quad (2)$$

Where $\Delta(\Delta T) = (T_{i+1} - T_i) - (T_i - T_{i-1}) = T_{i+1} - 2 T_i + T_{i-1}$

$\delta T = T_{j+1} - T_j$, with i indexing across the node and j indexing over time.

A numerical algorithm developed by Pafko [6] was adapted for use here. Written in Visual C++, thermal conductivity, density, & heat capacity can all be functions of temperature, time, and position. Appropriate values were inserted into the algorithm for CBT using data obtained by experiment and modelled using a simple curve fit (thermal conductivity) and from material data sheets and the literature for density and specific heat [7].

A fixed temperature boundary condition experiment was carried out to help verify the model. A stack of six CBT-RFI / glass fibre layers was inserted between two platens pre-heated to 120°C. The temperature profile through the thickness was recorded.

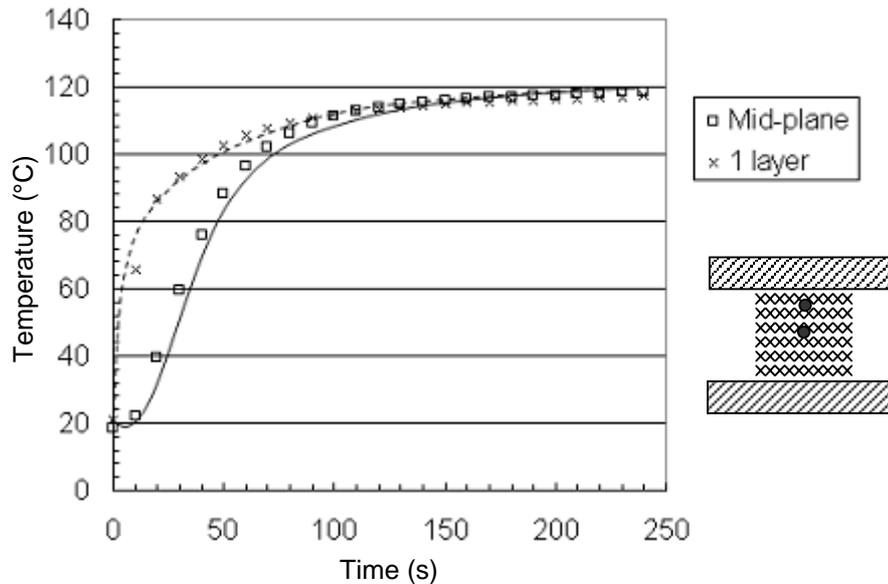


Figure 7 1-D experimental vs. model data

The experimental data is plotted against the model data in Figure 7. There appears to be good agreement for both the mid-point node and also at a node one layer in from the platen surface. As stated previously however, this model is only adequate for the initial heating stage up to 120°C and needs to be expanded to include phase change effects between 130-150°C, and subsequently, the heat transfer in the melt phase up to 195°C, the recommended polymerisation temperature for CBT.

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