Characterization of Velocity Profile of Highly-Filled GFRP-BMC through Rectangular Duct-Shaped Specimen during Injection Molding from SEM Fiber Orientation Mapping

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There is an extensive body of research and tests on numerical simulation to obtain rheological properties of injection molded resins and composites, however to our knowledge there is no or little research on estimating velocity profile from fiber orientation mapping of GFRP-BMCs (glass fiber reinforced polymer bulk molding compounds). This study reports a simple analysis of specific laminar creep flow properties of highly-filled short-fiber GFRP-BMC through a dogbone specimen as a rectangular duct during injection molding from SEM fiber orientation mapping which is found to exhibit a 3-layer [skin-core-skin] structure resembling classical laminar flow through a conduit. Therefore, to characterize the BMC with extremely low Reynolds number \( \sim 2 \times 10^{-4} \), velocity profile, primary and secondary boundary layers were estimated from Hermans fiber orientation parameter across gauge section thickness. (doi:10.2320/matertrans.M2013143)

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1. Introduction

Glass fiber reinforced polymer bulk molding compounds (GFRP-BMCs) are 3-phase systems composed of short glass fibers, polymer and contain a high \( \sim 35 \) to over 50 mass% of filler such as CaCO₃. Glass fiber reinforcement usually ranges from 5 to 30 mass% while glass fiber length ranges from about 3.2 to 12.7 mm (1/8 to 1/2 in.).¹,² Formulations are optimized for precise dimensional control, flame resistance, high dielectric strength, corrosion and stain resistance and color stability. BMCs are used for aerospace, automotive parts, housing for electrical wiring and corrosion-resistant needs, hence mechanical property improvement is essential for durability and use life. BMCs have excellent flow characteristics that make them well-suited for injection-molded parts requiring precise dimensions and detail.

There is an extensive body of research and tests on numerical simulation to obtain rheological properties of injection molded resin and their GFRPs dating back until at least the 1970s, several cited by Greene and Wilkes.³ However, to our knowledge there is no or little research on estimating velocity profile and its flow parameters from fiber orientation mapping. Some recent works on GFRP fiber orientation include microstructure characterization,⁴,⁵ interaction with processing parameters and mechanical properties,⁶-¹⁰ and mathematical modeling.¹¹ Two-phase fiber-polymer polyamide 6,6 exhibits a 5-layered flow pattern across specimen thickness [skin- shell- core -shell -skin] consisting of highly random thin skins approximately 4% of sample thickness with relatively thick highly oriented shell layers surrounding a thick inner core with a near random in-plane fiber orientation.⁹ In short and long glass fiber polyamide injection-molded rectangular cuboid-shaped plaques, Akay and Barkley correlated longitudinal and transverse fiber flow orientations with tensile and dynamic mechanical and fracture properties.¹⁰ Taking cross sections parallel to the flow line at various depths, they found a 3-layer [skin-core-skin] flow pattern appearing to transition into 5-layers with depth. Core thickness also increased with injection speed, melt and mold temperatures.¹⁰

A typical model used for injection molding simulation is by Hele-Shaw¹² that gives simplified equations for non-isothermal, non-Newtonian, inelastic flows in a thin cavity. Normally the following assumptions are proposed by Hele-Shaw prior to mathematical modeling: 1) cavity thickness is much less than width; 2) incompressible polymer melt behaves as viscous with no elastics where viscosity is shear-dominated; 3) viscous force is much higher than inertia or gravitational forces; 4) in thickness direction, velocity and pressure gradient are zero; 5) no fountain flow exits at flow front; 6) velocity is zero at mold walls; and 7) heat transfer between mold and paste is dominated by conduction then heat transfers into the core by convection. If we used the Hele-Shaw model, assumptions 1), 4) and 5) do not apply to the BMC since cross-sectional dimensions are 3.3 \( \times \) 12.6 mm; and fountain flow is observed at the flow front. While widely used, the Hele-Shaw model does not take into account fiber orientations with velocities, therefore the new approach of flow-front mechanics is proposed.

We do not claim to simulate the entire injection molding process. However, we claim it is possible velocity profile of the paste through the mold cavity and some rheological properties can be estimated from fiber orientation mapping with respect to mold flow direction, and shot time \( t_s \), it takes for the paste to fill the mold. This is demonstrated by SEM mapping of a cross section perpendicular to the mold flow across the specimen thickness at the center of the gauge section by meticulously positioning ellipses over 1,200 fiber cross sections with power point computer program to obtain fiber orientations. Since this took much time, only one cross section could be examined. This method could possibly be used as an additional evaluation to obtain a rough or better
mold flow direction

Fig. 1 Fiber orientation in spherical coordinates as a function of $\theta$ and $\phi$: angles (deg) with respect to the molding direction $z$-axis and width direction $y$-axis of sample, respectively.

This is assuming circular fiber cross sections.

Measurement accuracy of fiber cross-sections depends on computer screen size, resolution of computer program in positioning ellipses and resolution of SEM photomicrographs. For the conditions of this experiment, ellipses could be sized in 650 nm increments, therefore error was within $+/−650$ nm. Hence, the error in measuring ellipse short axis, $a$ (fiber diameter) was within $+/−5.7\%$ since average fiber diameter ($a_{avg}$) was measured to be 11.36 µm. Likewise, the average long axis of the ellipses, $b_{avg}$ was 23.19 µm, thus percent error for long axis was with $+/−2.8\%$.

Fiber angle orientation mapping shows the 3-phase GFRP-BMC has been found to exhibit a 3-layered flow pattern of [skin-core-skin] where the profile is inferred here as a solidification wavefront 3-D elliptical paraboloid resembling classical laminar flow through a conduit that can apparently be estimated by Navier-Stokes equations.

Picturing the mold cavity as a straight rectangular duct, there are factors to consider. First, the BMC is opaque and has low contrast between fiber and matrix. While the accepted transition Reynolds number ($Re$) from laminar to turbulent flow is $\sim2300$ for circular pipe, the thick paste 3-phase BMC injection-molded polymer composite exhibits extremely low $Re \sim2.4 \times 10^{-4}$ undergoing highly viscous laminar creep motion classified as Stokes Flow, and lower than 2-phase fiber-polymer systems without filler. Stokes Flow is typically accompanied by a thick boundary layer where viscous forces dominate over inertial forces.

These extremely low $Re$ numbers act as an advantage for homogeneity of the part since entrance length, $L_e$ at which the flow pattern and boundary layer stabilize calculated for laminar flow is nearly negligible in comparison to mold cavity length. Therefore, to characterize the GFRP-BMC flow through a conduit, the velocity profile and its flow parameters are estimated from fiber angle distribution across gauge section thickness.

2. Experimental

2.1 Preparation of GFRP

Premix, Inc. of North Kingsville, Ohio, provided an injection molded BMC (bulk molding compound) consisting of: 13.95 mass% propylene glycol maleate polyester (33 mass% styrene solution), 13.95 mass% styrene butadiene copolymer (70 mass% styrene solution), 20.00 mass% Owens and Corning OCF-405 E-glass fibers, 47.10 mass% calcium carbonate filler (CaCO$_3$), 0.35 mass% t-butyl perbenzoate, 0.10 mass% t-butyl peroctoate, 0.25 mass% carbon black pigment, 1.20 mass% calcium stearate, 3.00 mass% aluminum silicate and 0.10 mass% magnesium oxide.

To obtain flow parameter approximations taking into account the non-Newtonian shear thinning behavior of the BMC paste, dynamic viscosity measurements of Couette flow were carried out by rheometer at Premix, Inc. A linear Arrhenius relationship between dynamic viscosity $\eta$ (Pas) and shear rate $\dot{\gamma}$ ($s^{-1}$) was obtained at 20°C where viscosity was $9.0 \times 10^2 < \eta < 4.3 \times 10^3$ Pas for shear rates between $10^2 > \dot{\gamma} > 10^5 s^{-1}$. In the specimen, $\dot{\gamma}$ cannot be measured directly, but can be calculated from the measured average flow (bulk) velocity, $U_b$ by the Mooney-Rabinowitsch equation:

$$\dot{\gamma} = 4Q/\pi R^3 = 8U_b/D_H$$

where $\dot{\gamma}$ is referred to as “average shear rate”, $Q$ is volumetric flow rate ($m^3/s$), $R$ is radius if circular pipe (m), $U_b$ is bulk velocity (0.135 ms$^{-1}$), $D_H$ is hydraulic diameter (m), for a square duct equal to $4A/P$ where $A$ is specimen cross sectional area and $P$ is wetted perimeter calculated as total perimeter, $2(w + h)$, hence $\dot{\gamma}$ is $206 s^{-1}$. Interpolation of the Arrhenius relationship yields bulk viscosity $\eta_b$ of $5.0 \times 10^3$ Pas which we will use for calculations. The polyester melt viscosity, $\eta_{melt}$ is $\sim2$ Pas at 163°C.

Volume fractions, $V_f$ of the E-glass fibers, CaCO$_3$ filler and polymers with remaining components were 0.1317, 0.2902 and 0.5781, respectively.

Paste with 3.2 mm fibers was mixed in a double-arm sigma blade mixer for a total of 50 min at room temperature. The paste was allowed to stand for several hours and then injected molded into an ASTM D-638 family mold for dogbone-shaped specimens with a $3.03 \times 10^2$ N (75 ton) New Britain with the following processing parameters: mold temperature 436 K (163°C), barrel temperature $RT$, injection pressure 3.50–6.90 MPa, shot time, $t_s = \sim2.0$ s, hold time 15 s and cure time 1.5–2.0 min. Average fiber length, $l$ of about 1,000 fibers was measured to be 0.44 mm with a standard deviation of 0.203 mm by SEM. In general, two standard deviations comprise approximately 95% of the population, which is 0.04 mm $< l < 0.85$ mm.
Sample dimensions total length, gauge length, width and thickness were ~210, 100, 12.6, 3.3 mm, respectively, with cross-sectional area of 42.0 mm$^2$ in the gauge length.

2.2 Scanning electron microscopy (SEM)

A Jeol JSM-35CF scanning electron microscope (SEM) was used to observe polished surfaces of the composite. Specimens were polished with alumina polishing paper with successive finesses and sputtered with Pt. A mosaic of SEM photos across the specimen thickness was assembled.

3. Results

3.1 Composite specimen thickness

Figure 2 shows position of mapping of SEM photomicrographs across the 3.3 mm thickness for GFRP-BMC specimen perpendicular to mold flow direction z-axis. This was sectioned into 24 areas from 1 to 24 to obtain effect of position across thickness.

Figure 3 shows the mapping, which to our knowledge is the first time GFRP-BMC composites fiber orientation map is illustrated in the literature. Since the BMC is opaque and has low contrast between fiber and matrix, Fig. 3 is a mosaic of SEM photomicrographs put together where $\theta$ are measured by meticulously sizing and positioning ellipses over the fiber cross sections using powerpoint computer program. Mold flow direction is normal to the plane of the page. Across the 3.3 mm thickness fibers appear to have a fountain configuration, the center fibers appearing to have been pushed out perpendicular with respect to the flow at highest velocity decreasing near or to zero near the mold walls. Fiber density distribution is inhomogeneous. There are fiber-rich areas in most sections while notably sections 4–6 have low fiber density. Some fibers appear to have been bent in sections 6–8 or broken during the mixing and the injection molding process.

Figure 4 shows average measured angle, $\phi$ (deg) with respect to $y$-axis, parallel to specimen width for each section through thickness, $th$. Standard deviations (deg, in brackets) show variations in $\phi$ are wide due to the fountain flow nature of the BMC paste. Interestingly, the average $\phi$ do not deviate much from the width direction, showing the shorter dimension ($th = 3.3$ mm) controls, or squeezes the flow patterns. Note these standard deviations in angle $\phi$ do not designate the preciseness of the ellipse measurement, they merely show variation in flow. Ellipse measurements are quite accurate to $+/−650$ nm and are described in the introduction yielding $+/-5.7$ and $+/-2.8\%$ for ellipse short and long axes ($a$ and $b$), respectively.
Figure 5 shows an SEM micrograph at higher magnification near the mold wall where many fibers are highly oriented to the mold flow direction. The three marked fibers (lines) to the left have ratio of ellipse long and short axes \( b/a = 1.18, 1.26 \) and 1.21, hence \( \theta = 32.0, 37.2 \) and 34.6 deg, from mold flow direction respectively, while angle with respect to \( y \)-axis width direction, \( \phi = 4, -5 \) and \(-10 \) deg. The three marked fibers to the right have \( b/a = 1.016, 1.030 \) and 1.052, thus \( \theta = 10.4, 13.9 \) and 18.1 deg, while their \( \phi = -1, 2 \) and 0 deg, respectively. Figure 6 shows an area near the center of the specimen thickness where fibers are oriented more perpendicular to the mold flow direction with marked fibers from left to right having \( b/a = 5.24, 10.81 \) and 5.80, respectively, hence \( \theta = 79.0, 84.7 \) and 80.1 deg, with \( \phi = -13, -17 \) and \(-18 \) deg.

Figure 6 SEM micrograph showing fibers oriented more perpendicular to the mold flow direction near the center of the thickness.
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\[ U_{\text{max}} = 0.202 \text{ ms}^{-1} \]

\[ U_b = 0.180 \text{ ms}^{-1} \]

\[ U_0 = 0.135 \text{ ms}^{-1} \]

\[ U_c = 0.194 \text{ ms}^{-1} \]

\[ \rho_{(A)} = 498 \text{ mm}^{-2} \]

\[ \rho_{(A)} = 156 \text{ mm}^{-2} \]

\[ v_y = U_0 \left[ 1 - (2x/th)^2 \right] \]

\[ \rho_{(A),\text{avg}} = f(V_f) \]

\[ \rho_{(A),\text{calc}} = \rho_{(V_f),\text{avg}} \]

\[ \rho_{(A),\text{calc}} = \rho_{(V_f),\text{avg}} \]

\[ \delta_j = \sum_i [N(\theta_j) \cos^2(\theta_j)] / \sum_i [N(\theta_j)] \] (6)

where \( N(\theta_j) \) is number of fibers in each section, and \( i \) and \( j \) are fiber, and section number, respectively. Hence, velocity for each section, \( v_j \), is estimated from \( U_b \). To fit data to inverted parabola, orientation parameter is inverted (1 - \( \eta_{ij} \))avg in eq. (7):

\[ v_j = \frac{U_b}{(\sum_i [N(\eta_{ij})/(1 - \eta_{ij})]/24)}(1 - \eta_{ij}) \] (7)

where \( (1 - \eta_{ij})\text{avg} = (\sum_i [N(\eta_{ij})/(1 - \eta_{ij})]/24) \). Figure 8 shows the experimental data are well fit to a parabolic velocity profile in which the viscous displacement effect.18)

 Assigning primary boundary layer as that of flow, past research indicates it would be quite thick since as \( Re \) number decreases boundary layer thickness increases due to the large viscous displacement effect.18)

To estimate \( \delta \) for the GFRP-BMC flowing through the rectangular duct therefore, we follow the tradition of taking the points on the experimental curve of Fig. 9 that are 99% of the centerline velocity, 0.99\( U_c \).14) Graphical interpolation for designated left and right sides yields \( \delta_{0.99U_c,(L)} = 0.49 \) and 1.61 mm (\( \delta_{0.99U_c,(R)} = 1.05 \text{ mm} \)), respectively (Fig. 9) encompassing ~67% of the thickness. This is characteristic of the laminar creep flow of the highly viscous BMC.

In addition, we define a secondary boundary layer, \( \delta_t \) for fiber reinforced composites as the distance from the mold walls in which fibers remain highly oriented with the flow, agreeing with other composite systems.10,11) This is done simply from the “skins” in Fig. 9 as: \( \delta_{t,(L)} = 0.14 \) and 0.27 mm, respectively, hence \( \delta_{t,(avg)} = 0.20 \text{ mm} \). Here fibers are \( \theta = 0 \) to 20 deg with respect to the mold flow direction. This is apparent in the mosaic of Fig. 3.

4.3 Parabolic velocity profile fit of BMC paste flow from experimental values

To solve for the Navier-Stokes equations, the experimental data is fit to a parabolic velocity profile in which the sum of velocities, \( v_y \), must equal that from the experimental data. Figure 9 shows the plot of eq. (8):

\[ v_y = U_{c,\text{calc}}[1 - (2x/th)^2] \] (8)

where iteration yields calculated centerline velocity (\( U_{c,\text{calc}} \)) of 0.194 ms\(^{-1}\). Although the positions of the experimental maximum velocities are higher and lower than the calculated parabolic fit on left and right sides respectively of Fig. 9, the parabola appears to be a decent approximation of the 3-layer flow. The difference between the experimental and calculated velocity profiles can be explained by Fig. 10 where higher velocities above that of the parabolic fit are accompanied by lower fiber densities, \( \rho_{(A)} \) below the average (\( \rho_{(A),\text{avg}} \)) of 498 mm\(^{-2}\) (solid line) in sections 3 to 10. The low \( \rho_{(A)} \) appears

Fig. 8 Calculated velocity profile of GFRP-BMC flowing through gauge section of mold showing primary (\( \delta_{t,u} \)), and secondary (\( \delta_t \)) boundary layers for left (L) and right (R) specimen walls, respectively.

Fig. 9 Parabolic profile fit across the 3.3 mm specimen thickness.

Fig. 10 Measured fiber density, \( \rho_{(A)} \) perpendicular to molding direction for highly filled BMC paste. Area, \( \rho_{(A),\text{avg}} \) (solid line); average \( \rho_{(A)} \) (dotted line) are indicated.
to allow the paste to flow faster through the mold causing the sparse fibers to be pushed out at higher angles perpendicular to the flow as shown in the Fig. 3 exhibiting a fountain configuration. On the other hand, on the right side of Fig. 9 lower velocities are accompanied by higher \( \rho h_{\text{A}} \) above \( \rho h_{A_{\text{avg}}} \) in sections 12 to 23 causing higher resistance to flow and lower velocities than the parabolic fit.

For the parabolic fit, boundary conditions \( v_z = 0 \) are set at mold walls \( x = \pm L \). Figure 9 shows calculated primary boundary layers \( \delta_{0.99u_{c,\text{calc}}} \) are 1.50 mm (3.00 mm total) extending throughout most of the thickness.

4.4 Approximating effective viscosity, \( \eta_{\text{eff}} \) at mold walls by steady-state Navier-Stokes equation

Picturing the BMC paste as forming a thin melt layer with lower viscosity within \( \delta_t \) at the mold walls assisting to push the ultra-high viscosity core into the mold; it seems logical effective viscosity, \( \eta_{\text{eff}} \) at the mold walls can be estimated from injection pressure, \( \delta P \) using the Navier-Stokes equation. For flow along the z-axis,\(^{19}\)

\[
\rho g z - \frac{\delta P}{\delta z} + \eta (\delta^2 v_z / \delta x^2 + \delta^2 v_z / \delta y^2 + \delta^2 v_z / \delta z^2) = \rho (dv_z/dt) \tag{9}
\]

where the terms are: gravitational, pressure, viscous and acceleration forces per unit volume. Although the paste is a non-Newtonian shear thinning fluid\(^{20}\) and there are temperature gradients, this is beyond the scope of this study and seems to be taken into account in the frozen boundary layers, this is beyond the scope of this study and seems to be taken into account in the frozen fiber orientation.

Since the density of the thick BMC paste is approximately equal to that of the molded product because it contains 67.1 mass\% hard components of glass fiber (20 mass\%) and CaCO\(_3\) filler (47.1 mass\%) and only \(-28\) mass\% polymer, the Navier-Stokes equations are simplified as an incompressible Newtonian fluid with negligible gravitational effects eq. (9):

\[
\delta P / \delta z = \eta (\delta^2 v_z / \delta x^2 + \delta^2 v_z / \delta y^2) \tag{10}
\]

where the velocity profile for laminar flow is approximated as the 3-D elliptical paraboloid:

\[
v_z = U_{c,\text{calc}}[1 - (2x/\eta)^2 - (2y/\eta)^2] \tag{11}
\]

where \( U_{c,\text{calc}} \) is 0.194 ms\(^{-1}\). Substituting eq. (11) into eq. (10) 2nd order implicit differentiation yields:

\[
\delta P / \delta z = 8\eta U_{c,\text{calc}}[(1/\eta)^2 + (1/\eta)^2] \tag{12}
\]

The pressure drop across the three sections, ‘A’, ‘B’ and ‘C’ therefore is the sum:

\[
(\delta P / \delta z)_{\text{tot}} = \Sigma_{k} (\delta P / \delta z)_{k} \text{ (k goes from A to C)} \tag{13}
\]

Rearranging, we obtain effective viscosity, \( \eta_{\text{eff}} \) as a function of injection pressure, \( \delta P \):

\[
\eta_{\text{eff}} = (\delta P / \delta z)_{\text{tot}} / [8(2U_{c,\text{A}}(1/\eta)^2 + (1/w\text{A})^2] + U_{c,B}[(1/\eta\text{B})^2 + (1/w\text{B})^2] \tag{14}
\]

where \( U_{c,A} = U_{c,C} = 0.126 \text{ ms}^{-1}, \ U_{c,B} = 0.194 \text{ ms}^{-1} \) and \( \delta z = \text{total specimen length}, \ L \) (0.220 m). The plot in Fig. 11 shows the relation yielding effective viscosity range at mold walls estimated as 47.6 < \( \eta_{\text{eff}} < 93.9 \text{ Pas} \) (\( \eta_{\text{eff(avg)}} = 70.8 \text{ Pas} \)) that must be overcome by the 3.5 < \( \delta P < 6.9 \text{ MPa} \) injection pressures. Exceeding the polyester melt viscosity of \( \sim 2 \text{ Pas} \), this seems a decent approximation since additives of shrinkage control, cross-linking, thickening and mold release agents, with pigment and filler all play a role in increasing viscosity above the polymer melt.

4.5 Hydraulic head pressure loss and entrance length

Since the present definitions of entrance length, \( L_e \) and hydraulic head pressure loss, \( P_{\text{loss}} \) are functions of the friction factor, \( f \)\(^{11}\) which depends primarily on the flow conditions at the duct walls, \( \eta_{\text{eff}} = 70.8 \text{ Pas} \) is used. \( P_{\text{loss}} \) of the paste entering section “A” at the gate can be calculated by the Darcy-Weisbach equation valid for duct flows of any cross section for laminar and turbulent flow:\(^{18}\)

\[
P_{\text{loss}} = f(L/D_H)(\rho U_{c,\text{B}}^2)/2 \tag{15}
\]

where \( \rho \) is density of the paste. Friction coefficient for laminar flow calculated by \( f = 64/Re^{15,16,22} \) is 5420 hence, \( P_{\text{loss}} \) is 1.32 MPa. Thus, \( P_{\text{loss}} \) is calculated to be 19–38\% of the 3.50–6.90 MPa injection mold pressure.

In addition, entrance length at which the boundary layer stabilizes, \( L_e \) for laminar flow is\(^{13,15,16}\)

\[
L_e/D_H = 0.06 Re \tag{16}
\]

Using this equation with \( \eta_{\text{eff}} = 70.8 \text{ Pas} \), \( L_e \) is calculated to be only 0.0054 mm for entering either sections “A” or “B” (Fig. 7), (0.002–0.005% of length) indicating primary boundary layer, \( \delta_{0.99u_{c,\text{calc}}} \) and flow is approximately constant and stable throughout the entire 210 mm length. This calculation indicates a steady-state flow condition of the fibers probably contributing to the excellent flow characteristics of BMCs that make them well-suited for injection-molded parts requiring precise dimensions and detail.

5. Conclusions

In conclusion, velocity profile of highly-filled GFRP-BMC that exhibits the laminar creep flow through rectangular duct-shaped specimen during injection molding from SEM fiber orientation mapping has been investigated. To our knowledge there is no or little research on estimating velocity profile from fiber orientation mapping of GFRP-BMCs.
The BMC was found to exhibit a 3-layered [skin-core-skin] flow pattern that resembles classical laminar flow through a conduit and there was decent agreement with a parabolic curve fit. Variations from a perfect parabola were attributed to fiber density gradients across the specimen thickness. The subsequent Navier-Stokes calculation appeared to reliably estimate viscosity of melt layer at the mold walls.

Mapping apparently allowed primary and secondary boundary layer values for flow and fiber orientation, respectively to be obtained. Primary boundary layer for fiber flow was estimated to stabilize at only 0.002–0.005% of the specimen length from entrance length calculations explaining the excellent flow characteristics of BMCs. Secondary boundary layer for fiber orientation was found to agree with that of unfilled GFRPs at 4–8% of the thickness. The fiber orientation mapping appeared to provide specific information about fiber distributions and a decent approximation of flow parameters.

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