Supplemental Material for Monolithic Multilayer Microfluidics via Sacrificial Molding of 3D-Printed Isomalt

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Precision Dispensing

Free-form 3D printing as demonstrated in this paper was achieved using a novel feedback system for precision dispensing. Below is a summary of existing dispensing techniques and a technical description of our technique.

Filament Drives

The most common extrusion method among commercial 3D printers is a filament drive, in which a plastic filament, typically 1.5 mm or 3 mm in diameter, is forced through a die or nozzle by apposed gears. Filament drives, however, are incompatible with materials that cannot be processed into a filament or cannot withstand the crushing force exerted by the gears.

Air-Over Dispensing

Commercial systems for precision dispensing often use an “air-over” system, in which material contained in a syringe is forced through a nozzle by compressed gas. This has the advantage of allowing very rapid changes in pressure, and thus rapid stopping and starting of the material flow. However, air-over systems are not designed to be heated. The frequent cycling of the gas over the material in the syringe results in rapid heat loss, which makes precise temperature control difficult. Also, commercial systems typically use polypropylene syringes, which limits the maximum temperature to the melting point of polypropylene, which can be as low as 130 °C.

Screw Auger Extrusion

Single or twin screw extruders rely on shear force to generate pressure behind an extrusion die. These are very commonly used in polymer and food processing. For shear-sensitive materials, screw extruders are a poor choice. Also, turning of the screw does not result in immediate material flow, as there is a time constant associated with the elastic deformation of the material.

Positive-Displacement Extrusion

Positive-displacement extrusion can be implemented using a piston or progressive cavity pump. In principle, the amount of material dispensed by a positive-displacement system is a function of
plunger position, and independent of the material being dispensed. In practice, however, elastic seals, tubing, and air bubbles create a non-negligible amount of compliance within the system. The effect can be modeled as a low-pass filter as shown in Figure S1.

$$\frac{Q_p(s)}{Q_n(s)} = \frac{1}{1 + sRC}$$

*Figure S1. Schematic of a positive displacement dispenser. $Q_p$ is the flow through the nozzle, $Q_c$ is the flow into the system’s “capacitance,” and $Q_n$ is flow through the nozzle. Flow is opposed by shear stress, denoted by $R$.*

Capacitance is determined largely by the presence of trapped gas, and is difficult to eliminate completely. Resistance is determined primarily by the viscosity of the fluid and the size of the nozzle. As shown below, this time constant can be on the order of minutes, preventing crisp stopping and starting of material flow. Thus, positive displacement is unsuitable for a high-fidelity printing.
Figure S2. Pressure decay over time using positive displacement to extrude molten isomalt through a 50-μm nozzle

**Positive Displacement with Pressure Feedback**

Having failed to identify a suitable commercial solution for precision extrusion of molten materials, we chose to develop a new method. This method uses a piston style positive-displacement extruder controlled using a pressure feedback measured near the nozzle. Controlling pressure instead of piston translation allows flow rate to be changed very rapidly. The control algorithm is enabled by the autofocus feature of the motion controller, which allows motor positioning using an analog signal for feedback.
Figure S3. Autofocus control loop for the Aerotech A3200 motion controller

The motion controller allows the feedback terms shown above. For this application, assume elastic compliance, such that

\[ \Delta P = k \Delta X \]

If \( \Delta P \) is the error between the current pressure and the desired pressure, and \( \Delta X \) is the resulting change in the position command, we have the control law

\[ \Delta X = \frac{1}{k} \times error \]

In z-space,

\[ \Delta X = X(1 - z^{-1}) = \frac{1}{k} \times error \]

\[ X = \frac{1}{1 - z^{-1}} \times \frac{1}{k} \times error \]

\[ X = \frac{z}{z - 1} \times \frac{1}{k} \times error \]

If we let \( \frac{1}{k} = Ki \), we see that

\[ X = \frac{z}{z - 1} \times Ki \times error \]
We should use the motion controller’s $K_i$ term exclusively and set the other gains to zero. $K_i$ is inversely proportional to the stiffness, or directly proportional to the compliance, of the system. This is in agreement with the intuitive fact that an increase in system compliance, for example, by introduction of an air bubble, should require greater movement of the piston in order to reach the target pressure.

This control law was implemented and manually tuned. As shown below, response time for a 100-psi step was less than 250 ms.

Figure S4. Step response for hybrid dispenser

In principle, this system could be implemented using a screw-type extruder or progressive cavity pump. It is also possible to design the system to control speed, rather than position.
Material Characterization

Because the mold is freestanding, the material must be sufficiently stiff that bending is negligible; barring that, it is desirable to measure the elastic modulus so that any bending can be modeled and the design modified to compensate for it.

![Four-point bending test setup](image)

*Figure S5. Four-point bending test setup*

We wish to determine the flexural modulus $E$ to facilitate modeling and comparison with other materials. For $L_i = L/2$, the deflection of the beam at the point of the load application is given by

$$\delta = \frac{FL^3}{96EI}$$

where $I$ is the moment of inertia of the beam cross section.$^{33}$ The plot of load against displacement is linear:

$$F = m\delta = \frac{96EI}{L^3}\delta$$

So, the modulus is given by
$E = \frac{mL^3}{96I}$

Mechanical testing using a 4-point bend apparatus was performed on 9 samples of isomalt with dimensions 9.525 × 9.525 × 70 mm. The distance between the supports was 60 mm, and the distance between the points of load application was 30 mm.

A representative trace is shown below. The step decreases in load are due to the brittle beam cracking under the contact stresses at the supports. This prevented measurement of the yield stress but still allowed for measurement of the modulus, which was based upon the slope of the first linear portion of the curve before cracking occurred.

**Figure S6.** Sample plot of load cell output as a function of deflection. Only the red portion is used to determine the modulus.
Table 1. Moduli of isomalt beams.

<table>
<thead>
<tr>
<th>Trial</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
<th>9</th>
</tr>
</thead>
<tbody>
<tr>
<td>Modulus (GPa)</td>
<td>2.0</td>
<td>2.4</td>
<td>2.4</td>
<td>2.5</td>
<td>3.2</td>
<td>3.2</td>
<td>2.4</td>
<td>4.1</td>
<td>1.9</td>
</tr>
</tbody>
</table>

The average value of the modulus was 2.6 GPa and the standard deviation was 0.59 GPa. This modulus is similar to that of engineering plastics and is sufficient for the construction of free-standing constructs.