

**Titre:** Identification of constitutive theory parameters using a tensile machine for deposited filaments of microcrystalline ink by the direct-write method  
**Title:**

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**Date:** 2009

**Type:** Article de revue / Journal article

**Référence:** Lourdel, N., Therriault, D. & Lévesque, M. (2009). Identification of constitutive theory parameters using a tensile machine for deposited filaments of microcrystalline ink by the direct-write method. *Journal of Micromechanics and Microengineering*, 19(9), 095017. doi: [10.1088/0960-1317/19/9/095017](https://doi.org/10.1088/0960-1317/19/9/095017)  
**Citation:**

 **Document en libre accès dans PolyPublie**  
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**URL de PolyPublie:** <https://publications.polymtl.ca/10381/>  
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**Version:** Version finale avant publication / Accepted version  
Révisé par les pairs / Refereed

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 **Document publié chez l'éditeur officiel**  
Document issued by the official publisher

**Titre de la revue:** Journal of Micromechanics and Microengineering (vol. 19, no 9)  
**Journal Title:**

**Maison d'édition:** IOP Publishing Ltd  
**Publisher:**

**URL officiel:** <https://doi.org/10.1088/0960-1317/19/9/095017>  
**Official URL:**

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1 Identification of constitutive theory parameters using  
2 a tensile machine for deposited filaments of  
3 microcrystalline ink by Direct-Write Method

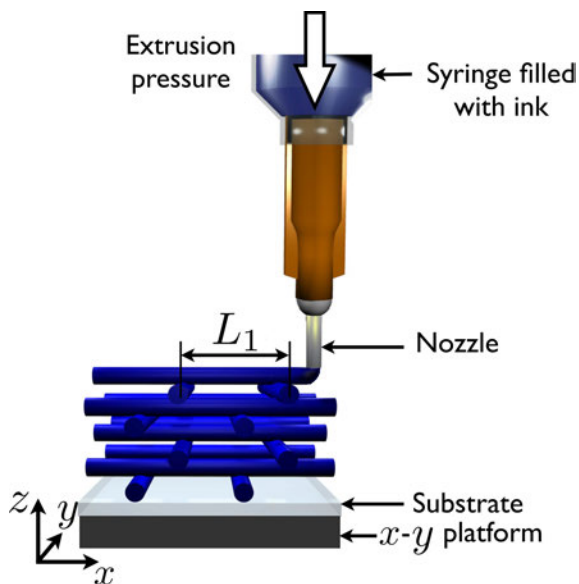
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9 **Abstract.** A custom-designed tensile machine is developed to characterize the  
10 mechanical properties of ink micro-filaments deposited by Direct-Write method. The  
11 Direct-Write method has been used for the fabrication of a wide variety of micro-  
12 systems such as microvascular networks, chaotic mixers and laboratory on-chips. The  
13 tensile machine was used to measure the induced force in ink filaments during tensile  
14 and tension-relaxation tests as a function of the applied strain rate, the ink composition  
15 and the filament diameter. Experimental data was fitted by a linearly viscoelastic  
16 model using a data reduction procedure in order to identify the constitutive theory  
17 parameters of the deposited ink filaments. The model predictions based on the defined  
18 constitutive theory parameters were closed to the experimental data generated in this  
19 study. Such models will be useful in the development and optimization of future 3D  
20 complex structures made by direct-write method.

21 Submitted to: *J. Micromech. Microeng.*



**Figure 1.** Schematic representation of a scaffold robotic deposition. Ink stored in a syringe is extruded through a nozzle under a constant pressure and deposited on a substrate in a defined pattern by adjusting the  $x$ - $y$  platform displacements and the nozzle height along the  $z$ -axis. Successive layers are deposited by incrementing the nozzle height and filaments are in suspension on the previous layer with a spanning distance  $L_1$ .

## 22 1. Introduction

23 Interactions with micro and nanochemical or biological entities have been improved with  
 24 the recent advancements in microfluidic, microelectronic and microelectromechanic de-  
 25 vices [1, 2, 3, 4]. For example, microelectronic devices are technically able to detect  
 26 or measure adhesion and proliferation of specific pathogenic biomolecules [3, 4]. These  
 27 advancements imply the use of new fabrication processes capable of dealing with more  
 28 complex structures like the Direct-Write Method (DWM) which corresponds to processes  
 29 that employ a deposition nozzle to extrude or deposite materials and a translation stage  
 30 to create a controlled pattern on substrates or devices [5]. Robotic deposition is one of  
 31 the DWM processes adapted for the deposition of various solid and soft materials in a  
 32 two-dimensional (2D) pattern or a three-dimensional (3D) scaffold [6]. This scaffold is  
 33 the first step of the fabrication process for micro-devices such as microfluidic mixers,  
 34 drugs delivery systems [7], micro fuel cells [8] and heat exchangers for cooling system [9].

35  
 36 Figure 1 shows a 3D scaffold fabrication with a viscoelastic ink using robotic de-  
 37 position process [10]. First, a patterned layer is deposited on a substrate by moving a  
 38  $x$ - $y$  platform with adjusted extrusion nozzle height, diameter and deposition pressure.  
 39 Then, extrusion nozzle height is incremented along the  $z$ -axis and successive layers are  
 40 deposited until the desired scaffold is obtained. Each layer is composed of ink filaments  
 41 supported by the previous layer with a defined spanning distance  $L_1$  and filaments

42 present a diameter closed to the inner diameter of the extrusion nozzle.

43

44 The fabrication of 3D scaffolds can be time-consuming, depending on the final  
 45 structure complexity ( $\sim 78$  minutes for a 104 layers scaffold consisted of a simple cubic  
 46 lattice of  $20\text{ mm} \times 20\text{ mm} \times 20\text{ mm}$  [10]). It is therefore of paramount importance that  
 47 inks are mechanically adapted to minimize 3D scaffolds alterations as a function of time.  
 48 Rheological studies led to the improvement of the inks viscoelastic properties [11, 12].  
 49 They are unable, however, to predict ink filament behaviour in scaffolds since they do  
 50 not take into account the fact that inks are extruded under a filament shape and that  
 51 the ink structure may be reorganized in the nozzle during extrusion [12]. Observations  
 52 of mid-span time deflection of spanning filaments have allowed the definition of the  
 53 minimum ink shear elastic modulus value allowed for limiting filament deflection in  
 54 scaffolds [13]. Observations have also led to the development of a structural model  
 55 for the time dependent deformation of a spanning ink filament simply supported at its  
 56 extremities and under its own weight [11]. Similarly, filament mid-span time deflection  
 57 observations are limited and can only be defined for a constant load corresponding to  
 58 the filament spanning weight. In this paper, we present a tensile machine especially  
 59 developed to characterize the mechanical behaviour of spanning ink filaments as those  
 60 robotically deposited in scaffolds. This machine allows imposing various load levels as  
 61 well as different strain rates. We also propose a data reduction procedure for obtaining  
 62 the parameters of a linearly viscoelastic constitutive theory that best fit experimental  
 63 data obtained from tensile tests.

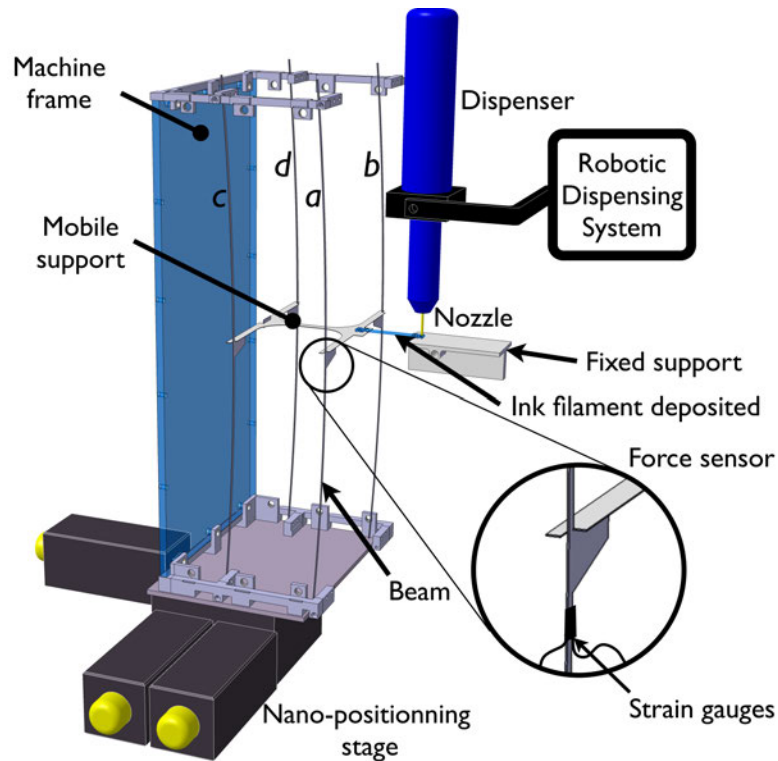
## 64 2. Material, experimental method and data reduction procedure

### 65 2.1. Material

66 The organic ink is a mixture obtained by melting ( $\sim 80\text{ }^\circ\text{C}$ ) and mixing ( $\sim 20$  minutes  
 67 by magnetic stirring) petroleum jelly (*USP grade Vaseline<sup>®</sup>, Leverpond's Inc., Toronto,*  
 68 *ON*) with a defined amount of microcrystalline wax (*SP18, Strahl & Pitsch Inc., West*  
 69 *Babylon, NY*). At the end of the mixing period, the ink is poured in a syringe and  
 70 immersed in cold water to avoid phases separation [12]. Microcrystalline wax content  
 71 is typically between 10 to 40 weight percent (wt.-%) of the ink to allow the deposition  
 72 of a 3D scaffold [10, 12].

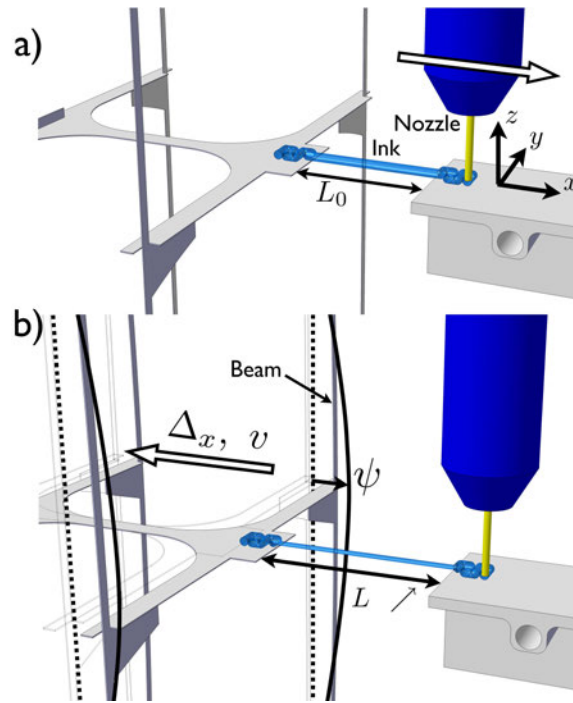
### 73 2.2. Experimental Method

74 The tensile machine is represented in Figure 2 and is used to measure the mechani-  
 75 cal properties of a robotically deposited ink filament spanning between a mobile and a  
 76 fixed support. The mobile support is attached to four thin beams ( $a - d$ ) clamped near  
 77 their extremities in a machine frame. The frame is held by a MAX341 nano-positioning  
 78 stage (*Thorlabs Inc., Newton, NJ*) able to manage 3-axis displacements with a 20 nm  
 79 resolution using stepper motors and an APT Modular Rack controller (*Thorlabs Inc.,*



**Figure 2.** Schematic representation of the tensile machine used to measure the mechanical properties of a robotically deposited ink filament spanning between a mobile and a fixed support. The mobile support lays on four beams ( $a$ - $d$ ) clamped to the machine frame, held by a nano-positioning stage. Strain gauges placed on both sides of beams  $a$  and  $b$  are used to create a force sensor.

80 *Newton, NJ*). A 2 inch LT3 3-axis Travel Translation Stage (*Thorlabs Inc., Newton,*  
 81 *NJ*) (not shown) is used to manually adjust the position of the fixed support (dis-  
 82 placement resolution  $\sim 7 \mu\text{m}$  along the three axes) in order to keep both supports top  
 83 surfaces aligned and to set the desired distance between them, called spanning distance  
 84  $L_0$ . Two strain gauges EP-XX-031DE-120 (*Vishay Intertechnology Inc., Malvern, PA*)  
 85 placed on both sides of the beams  $a$  and  $b$  near their middle and oriented along the  
 86 beams longitudinal axes are used to create a force sensor. After a calibration procedure,  
 87 the measured longitudinal strains of beams  $a$  and  $b$  can be related to the applied force  
 88 on the mobile support. For each beam ( $a$  and  $b$ ), strain gauges are linked together to a  
 89 Wheatstone half-bridge of a NI SC-2043-SG chipset (*National Instruments, Austin, TX*)  
 90 connected to a NI PCI-6221 data acquisition card (*National Instruments, Austin, TX*).  
 91 The measurements are then filtered using an exponential moving average, monitored  
 92 and saved with a LabView program (*National Instruments, Austin, TX*). Exponential  
 93 moving average is a common technique for filtering measurements obtained with sensors  
 94 and is calculated, here, with a set of data acquired during 0.5 s. An ink filament can  
 95 be extruded and deposited between these two supports using a I&J2200-4 Robotic Dis-  
 96 pensing System (*I&J Fisnar Inc., Fair Lawn, NJ*), an Ultimius 2400 pressure regulator  
 97 (*EFD Inc., Westlake, OH*) and a HP7x air powered dispenser (*EFD Inc., Westlake,*



**Figure 3.** Schematic representation of a) the ink filament deposition along the  $x$ -axis between the two supports with a defined spanning distance  $L_0$  and b) the ink filament tension along the  $x$ -axis due to the mobile support displacement  $\Delta_x$  and the velocity  $v$ . The mobile support displacement increases the spanning distance  $L$  and induces ink filament inner forces due to its rigidity, which tends to displace the mobile support from  $\psi$  and to deform the four beams.

98 *OH*). The air powered dispenser contains a syringe filled with ink and, coupled with a  
 99 general purpose micro-nozzle (*EFD Inc., Westlake, OH*), allows the deposition of ink  
 100 filament in suspension between the two supports. Finally, the tensile machine is placed  
 101 in a room at constant temperature near 21.5 °C.

102

103 Figure 3 shows the two main steps of the tensile test procedure. In the first step  
 104 (Figure 3.a), an ink filament is extruded and deposited along the  $x$ -axis from the mobile  
 105 support to the fixed support with a defined spanning distance  $L_0$ . The extrusion pres-  
 106 sure and deposition velocity must be appropriate to connect a spanning filament to the  
 107 two supports. Contacts between supports and ink filament have been checked to ensure  
 108 that they present enough friction to avoid ink slipping using a digital video camera  
 109 Evolution VF FAST Color 12-bit (*Media Cybernetics, Bethesda, MD*) with a resolution  
 110 near 15  $\mu\text{m}$  per pixel. In a second step (Figure 3.b), few seconds after the deposition  
 111 of the filament, the nano-positioning stage is used to move the mobile support of  $\Delta_x$   
 112 along the  $x$ -axis at the desired velocity  $v$ . By moving the mobile support, the spanning  
 113 distance  $L$  increases and the ink filament is stretched. The induced force  $F$  due to  
 114 ink filament tension tends to displace the mobile support of  $\psi$  along the  $x$ -axis and to  
 115 deform the four thin beams. Therefore, induced force  $F$  during ink filament tensile test

116 can be deduced using the calibrated strain gauges measurements.

117

118 During the calibration procedure, different masses were successively applied on the  
 119 mobile support using a pulley system. The gauge measurements demonstrated a linear  
 120 response as a function of the applied force  $F_{\text{applied}}$  with a resolution of approximately 20  
 121  $\mu\text{N}$  according to the ASTM E4-03 Standard and a maximum load capacity near 1 N. It  
 122 was also demonstrated that the force sensor presents a response time near 1 s and an  
 123 percent error equal to 3 % for an approximately applied force  $F_{\text{applied}}$  equal to  $4 \times 10^{-3}$   
 124 N.

### 125 2.3. Data reduction procedure

126 The data reduction procedure is based on a least square fit method between the exper-  
 127 imental induced stress  $\sigma$  and the theoretical induced stress  $\tilde{\sigma}$  for a linearly viscoelastic  
 128 ink filament being stretched.  $\sigma$  is equal to  $4F/\pi d^2$  where  $d$  is the filament diameter  
 129 adjusted from its initial value  $d_{\text{init.}}$  ( $\varepsilon = 0$  %) as a function of the applied strain  $\varepsilon$  and  
 130 the ink filament Poisson's ratio  $\nu$  ( $d = d_{\text{init.}}(1 - \nu\varepsilon)$ ). For all measurements,  $\nu$  is assumed  
 131 to be constant and equal to 0.5 [11]. The initial ink filament mean diameter,  $d_{\text{init.}}$ , is  
 132 measured at different positions along the spanning distance using a SZ61 stereomicro-  
 133 scope (*Olympus, Tokyo, Japan*) with a magnification of 3X and the Evolution VF FAST  
 134 camera.

135

136 For a one-dimensional (1D) linearly viscoelastic material, the theoretical induced  
 137 stress can be expressed as a function of the strain history by :

$$138 \quad \tilde{\sigma}(t) = \int_0^t C(t - \theta) \frac{d\varepsilon}{d\theta} d\theta \quad (1)$$

139 where  $C(t)$  and  $\varepsilon$  represent the relaxation modulus and the applied strain, respectively.

140 The relaxation modulus  $C(t)$  is expressed as

$$141 \quad C(t) = C' + \sum_{i=1}^n C_i \exp[-t/\lambda_i] \quad (2)$$

142 where  $C'$ ,  $C_i$  and  $\lambda_i \geq 0$  for thermodynamic stability [14].  $C'$  represents the fully relaxed  
 143 modulus (i.e., for  $t \rightarrow \infty$ ). The  $C_i$  and  $\lambda_i$  define the relaxation modes and  $n$  represents  
 144 the number of relaxation modes. The instantaneous modulus (i.e., for  $t \rightarrow 0$ ) is given  
 145 by  $C' + \sum_{i=1}^n C_i$ . In the case of a tensile test conducted at a constant strain rate  $\dot{\varepsilon}_0$ ,  $\tilde{\sigma}$   
 146 is given by :

$$147 \quad \tilde{\sigma}(t) = C' t \dot{\varepsilon}_0 + \dot{\varepsilon}_0 \sum_{i=1}^n \lambda_i C_i \left( 1 - \exp[-t/\lambda_i] \right) \quad (3)$$

148 Then, if the strain is held constant after  $t_0$  (i.e., a relaxation period), according to  
 149 Boltzmann's superposition principle,  $\tilde{\sigma}$  is given by :

$$\tilde{\sigma}(t) = C' t_0 \dot{\varepsilon}_0 + \dot{\varepsilon}_0 \sum_{i=1}^n \lambda_i C_i \left( \exp[-(t - t_0)/\lambda_i] - \exp[-t/\lambda_i] \right) \quad (4)$$

for  $t \geq t_0$ .

152

The least square problem used for obtaining  $C'$ ,  $C_i$  and  $\lambda_i$  is defined as [15, 16] :

$$\inf_{C', C_i \geq 0} R^2 = \inf \sum_{j=1}^m \alpha_j \left[ \sigma(t_j) - \tilde{\sigma}(t_j) \right]^2 \quad (5)$$

where  $R^2$ ,  $m$  and  $\alpha_j$  are the residual value, the number of experimental data points and the weighting factors for each square value  $[\sigma(t_j) - \tilde{\sigma}(t_j)]^2$ , respectively. In the first step, the data reduction procedure is executed by fixing  $n$  and the  $\lambda_i$  a priori. Typically  $n = 10$  was used and the  $\lambda_i$  were distributed on a log scale. In the second step, Equation (5) is solved and the  $C_i$  identified values that are much smaller than the others are removed. New  $\lambda_i$  are introduced around the remaining  $\lambda_i$  corresponding to the non-negligible  $C_i$ . In the third step, Equation (5) is executed anew and the whole procedure is repeated until  $R^2$  does not vary significantly. At the end, constitutive theory parameters  $C'$  and  $\{C_i, \lambda_i\}$  are obtained.  $\alpha_j$  can be adjusted when solving Equation (5) for several responses simultaneously, for which the number of data points might not be equal. This is detailed in Section 3.5 . The data reduction procedure is programmed and executed with the Global Optimization Toolbox of Maple 12 Software (*Maplesoft, Waterloo, ON*) and the algorithm employed is a local search.

### 3. Results and Discussions

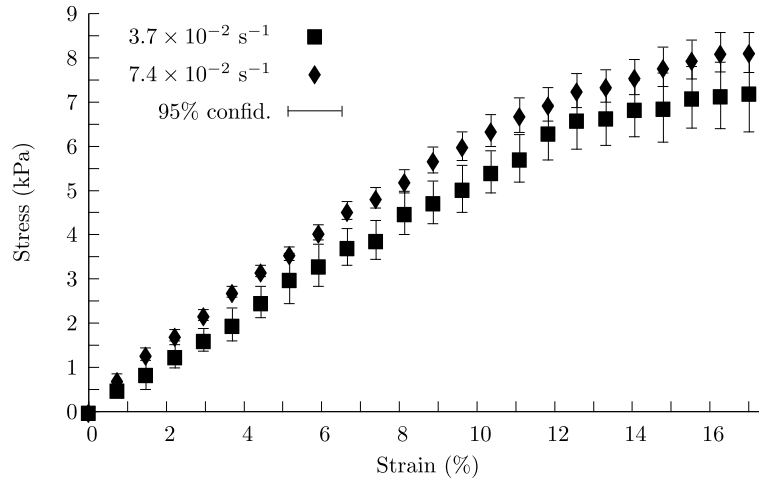
Ink filament response to tensile test is measured for different strain rates ( $\dot{\varepsilon}_0 = v / L_0$ ), microcrystalline wax amounts and extrusion nozzle inner diameters. The response is also measured for a tensile test with a defined strain rate  $\dot{\varepsilon}_0$ , followed by a relaxation test at  $t = t_0$  with a constant strain  $\varepsilon_1 = \dot{\varepsilon}_0 t_0$ .

173

At least 6 repetitions were conducted on different ink filaments for each parameters combination and each repetition respected the ink filament rupture requirements defined in the ASTM Standard C1557-03<sup>E1</sup>. The 95 % confidence intervals on the mean value are given for most of the measurements and only induced stress responses mean values are represented. The results are given for the interval  $t \in [0, t_f]$ , where  $t = 0$  and  $t = t_f$  correspond to the beginning of the tensile test and to the average ink filament rupture (characterized by the beginning of a decreasing slope of the induced stress response as a function of time). For the tension-relaxation test, measurements are stopped when acquired signals reach a stabilized value.

183





**Figure 4.** Evolution of the ink filament induced stress response as a function of applied strain during tensile tests conducted with two different strain rates,  $\dot{\epsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$  and  $7.4 \times 10^{-2} \text{ s}^{-1}$  on 40 wt.-% ink filaments extruded with a 0.84 mm nozzle.

184 For all experiments,  $L_0 = 10.67 \text{ mm}$  and tensile tests are mostly applied with  $\dot{\epsilon}_0 =$   
 185  $3.7 \times 10^{-2} \text{ s}^{-1}$ , which is a compromise between too low strain rates implying measured  
 186 induced force  $F$  in the same order of magnitude than the force sensor resolution in the  
 187 time interval  $t \in [0, t_f]$  and too important strain rates leading to ink filaments premature  
 188 failures.

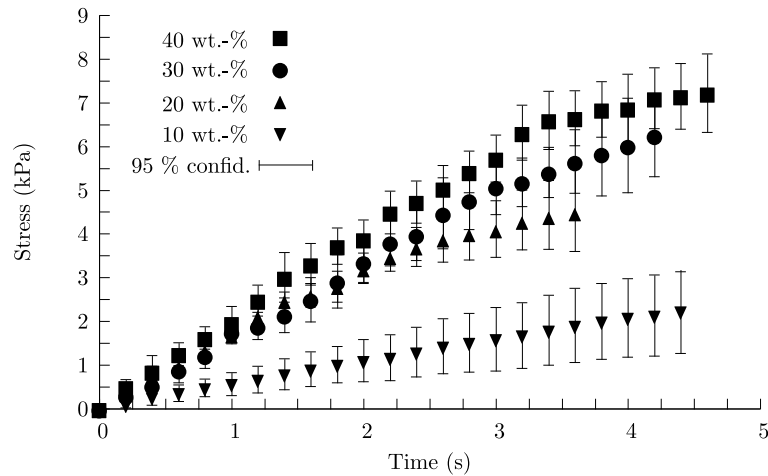
### 189 3.1. Ink filament response as a function of applied strain rate $\dot{\epsilon}_0$

190 Figure 4 shows the induced stress response  $\sigma$  as a function of the applied strain  $\varepsilon$  during  
 191 tensile tests conducted on 40 wt.-% ink filaments extruded with a 0.84 mm nozzle for  
 192  $\dot{\epsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$  ( $v = 0.4 \text{ mm.s}^{-1}$ ) and  $7.4 \times 10^{-2} \text{ s}^{-1}$  ( $v = 0.8 \text{ mm.s}^{-1}$ ). Ruptures  
 193 occur near  $\varepsilon = 16.5 \%$  for both strain rates and the material appears to be stiffer as the  
 194 strain rate is increased, suggesting that ink filaments are viscoelastic.

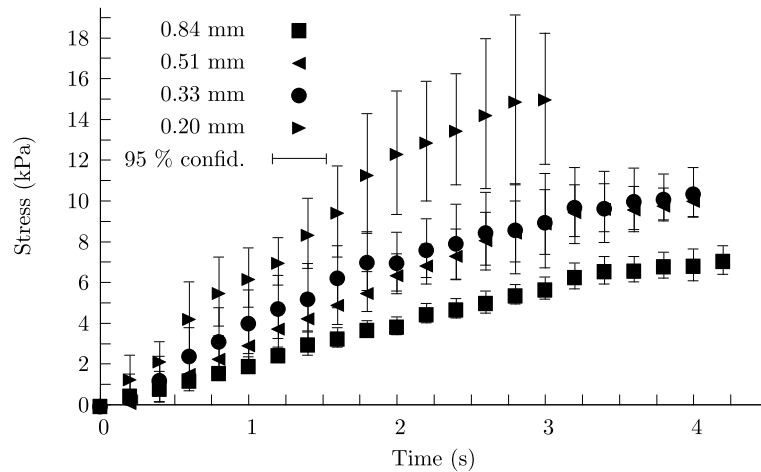
### 195 3.2. Ink filament response as a function of the microcrystalline wax amount

196 Figure 5 shows the induced stress response  $\sigma$  as a function of time obtained during  
 197 tensile tests with  $\dot{\epsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$  conducted on ink filaments with different amounts  
 198 of microcrystalline wax (10, 20, 30 and 40 wt.-%) and extruded with a 0.84 mm nozzle.  
 199

200 The induced stress response tends to increase with the amount of microcrystalline  
 201 wax and the response in the case of the 20, 30 and 40 wt.-% ink filaments is 200 %  
 202 or 300 % the value of the response of the 10 wt.-% ink filament. 20 and 30 wt.-%  
 203 ink filament responses are similar and their 95 % confidence intervals overlap for  $t <$   
 204 3 s. Although the 20 wt.-% ink filament presents an instantaneous modulus higher  
 205 than the 30 wt.-% ink filament for the same reason, instantaneous elastic modulus,  
 206 defined as  $E(t) = \sigma(t)/\varepsilon(t)$  with  $t$  near  $0^+$ , tends to increase with the microcrystalline



**Figure 5.** Evolution of the induced stress response as a function of time during tensile tests with  $\dot{\epsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$  conducted on ink filaments with four different amounts of microcrystalline wax : 10, 20, 30 and 40 wt.-%, and extruded with a 0.84 mm nozzle.

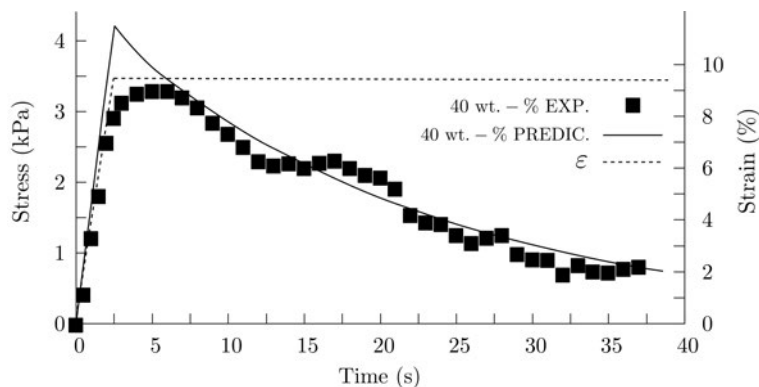


**Figure 6.** Evolution of the 40 wt.-% ink filament induced stress response as a function of time during tensile tests with  $\dot{\epsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$  conducted on filaments extruded with four different nozzle inner diameters : 0.84, 0.51, 0.33 and 0.20 mm.

207 wax content as shown in previous rheological studies [10, 12]. Instantaneous relaxation  
 208 moduli are approximately equal to 16, 44, 41 and 53 kPa for the 10, 20, 30 and 40  
 209 wt.-% ink filaments. Finally, the 20 wt.-% ink filament presents a rupture around 3.6 s  
 210 ( $\epsilon \sim 13.3 \%$ ) in comparison to the 10, 30 and 40 wt.-% ink filaments breaking between  
 211 3.9 ( $\epsilon \sim 14.5 \%$ ) and 4.5 s ( $\epsilon \sim 16.7 \%$ ), supposedly due to inaccurate deposition  
 212 parameters interfering with the mechanical behaviour of the 20 wt.-% ink filament.

### 213 3.3. Ink filament response as a function of extrusion nozzle inner diameter

214 Figure 6 shows the induced stress response  $\sigma$  as a function of time obtained during  
 215 tensile tests with  $\dot{\epsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$  conducted on 40 wt.-% ink filaments extruded  
 216 with different nozzle inner diameters (0.84, 0.51, 0.33 and 0.20 mm).



**Figure 7.** Evolution of the experimental induced stress response (40 wt.-% EXP.) of a 40 wt.-% ink filament extruded with a 0.84 mm nozzle as a function of time during a tensile ( $\dot{\varepsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$ ) and a relaxation ( $\varepsilon_1 = 9.3 \%$ ) test. The predicted response (40 wt.-% PREDIC.) based on the identified constitutive theory parameters is also shown.

217

218 The induced stress increases with the reduction of the nozzle inner diameter,  
 219 although responses for the 0.33 and the 0.51 mm nozzles are similar. The response  
 220 for the ink filament extruded with a 0.20 mm nozzle is nearly 200 % the responses with  
 221 the 0.33 mm and 0.51 mm nozzles and nearly 300 % the response with the 0.84 mm  
 222 nozzle. This increase may be explained by a microcrystalline alignment in the nozzle flow  
 223 direction with a reduction of the nozzle inner diameter. This alignment might increase  
 224 the ink filament stiffness along the extrusion (and testing) direction. In addition, the  
 225 ink filament rupture occurs earlier with the diminution of the inner diameter of the  
 226 nozzle. The rupture successively happens at  $t = 3 \text{ s}$  ( $\varepsilon = 11.1 \%$ ) for the ink filament  
 227 extruded with 0.20 mm nozzle, near 4 s ( $\varepsilon = 14.8 \%$ ) with the 0.51 mm and 0.33 mm  
 228 nozzles and 4.5 s ( $\varepsilon = 16.7 \%$ ) in the case of the 0.84 mm nozzle. These successive  
 229 ruptures confort the idea of a microcrystalline structure alignment phenomenon with  
 230 a reduction of the nozzle inner diameter implying a decrease of the filament ductility.  
 231 Finally, the reduction of the nozzle inner diameter leads to an increase of the 95 %  
 232 confidence intervals. This might be due to the difficulty to align exactly ink filaments  
 233 with the smaller diameters along the  $x$ -axis, which decreases the force measurements  
 234 accuracy [17]. This also may be due to the measured force  $F$  for the filament extruded  
 235 with the 0.20 mm nozzle which is only 20 % the force  $F$  measured in the case of the  
 236 0.84 mm nozzle and closer to the resolution of the force sensor.

### 237 3.4. Ink filament response to a tension-relaxation test

238 Figure 7 shows the induced stress response obtained during a tension-relaxation test  
 239 conducted on a 40 wt.-% ink filament extruded with a 0.84 mm nozzle. The tensile  
 240 test is conducted on the ink filament with  $\dot{\varepsilon}_0 = 3.7 \times 10^{-2} \text{ s}^{-1}$  and stops at  $t_0 = 2.5$   
 241 s. The relaxation part consists of applying a constant strain  $\varepsilon_1$  of 9.3 %. The ink

**Table 1.** Constitutive theory parameters  $C'$  and  $\{C_i, \lambda_i\}$  identified for different ink filaments with distinct combination of microcrystalline wax amount and inner diameter of extrusion nozzle.

Microcrystalline wax (wt.-%)	Extrusion nozzle (mm)	$C'$ (kPa)	$i$	$C_i$ (kPa)	$\lambda_i$ (s)
10	0.84	$\sim 0$	1	16.19	13.0
20	0.84	$\sim 0$	1	55.70	3.5
30	0.84	$\sim 0$	1	49.19	11.8
40	0.84	$\sim 0$	1	4.24	1.0
			2	46.05	21.0
40	0.51	$\sim 0$	1	96.61	6.65
40	0.33	$\sim 0$	1	132.41	2.85
40	0.20	$\sim 0$	1	196.76	4.3

242 filament presents a quasi-linear induced stress response during the tension test and an  
 243 exponential response with respect to time during the relaxation test. For  $t > 32$  s,  
 244 the stress response is fluctuating around 0.8 kPa and measurements are stopped at  $t$   
 245 = 37.5 s where the relaxation modulus i.e.,  $\sigma(37.5)/\varepsilon(37.5)$ , is approximately equal to  
 246 9.3 kPa. The 95 % confidence intervals are not shown in order to keep a legible response.

247

248 For this particular strain history, the theoretical induced stress response in a  
 249 viscoelastic material obeying Equation (2) should be maximum at  $t = t_0$  and decreasing  
 250 at  $t_0^+$ . However, the response reaches its maximum near  $t = 5$  s and starts decreasing  
 251 at  $t = 6.5$  s. This  $\sim 3$  s delayed response is supposed to be due to the response time of  
 252 the sensor for measuring successively increasing and decreasing forces.

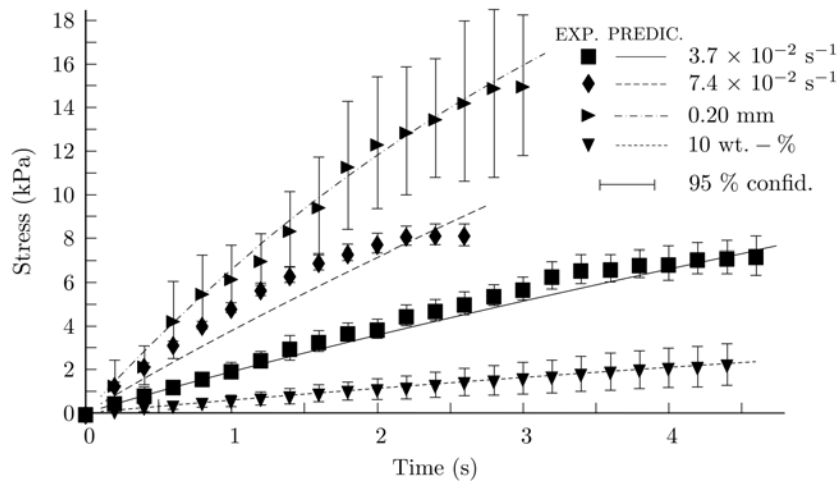
### 253 3.5. Identification of the constitutive theory parameters

254 Identification of the constitutive theory parameters  $C'$  and  $\{C_i, \lambda_i\}$  is conducted for  
 255 distinct ink filaments. Due to the previous mechanical observations, it is assumed here  
 256 that a distinct ink filament corresponds to a combination of a certain microcrystalline  
 257 wax content and a specific nozzle inner diameter. For each material, all experimental  
 258 data has been used in the definition of  $R^2$  from Equation (5). For example, in the case  
 259 of the 40 wt.-% ink filament extruded with a 0.84 mm nozzle, experimental data of  
 260 Figures 4, 5 and 7 ( $3.7 \times 10^{-2} \text{ s}^{-1}$ ,  $7.4 \times 10^{-2} \text{ s}^{-1}$  and 40 wt.-% EXP.) has been used  
 261 to indentify the constitutive theory parameters.

262

263 Table 1 lists  $C'$  and  $\{C_i, \lambda_i\}$  for distinct ink filaments. For any combination of  
 264 microcrystalline wax amount and nozzle inner diameter, the ink filament exhibits a neg-  
 265 ligible stabilized response ( $C'$ ). Further tensile and tension-relaxation tests over longer  
 266 periods of time would be required for validating these values.

267



**Figure 8.** Evolution of the experimental induced stress response (EXP.) as a function of time during tensile tests conducted on the 10 and 40 wt.-% ink filaments (sections 3.2), the ink filament extruded with a 0.20 mm nozzle (section 3.3) and the ink filament stretched at  $\dot{\epsilon}_0 = 7.4 \times 10^{-2} \text{ s}^{-1}$  (section 3.1). Predicted responses (PREDIC.) based on identified constitutive theory parameters are also shown.

268 In order to study the reliability of the identified parameters, Figure 8 shows, as  
 269 an example, the experimental data (EXP.) measured for different materials i.e., 10 and  
 270 40 wt.-% ink filaments extruded with a 0.84 mm nozzle (sections 3.1 and 3.2) and the  
 271 40 wt.-% ink filament extruded with a 0.20 mm nozzle (section 3.3), with their cor-  
 272 responding predicted responses (PREDIC.) based on the identified constitutive theory  
 273 parameters. The predicted responses are closed to the experimental data i.e., in the  
 274 95% confidence intervals, for the 10 wt.-% ink and the 0.20 mm nozzle ink filaments.  
 275 Similar prediction results have been obtained for the other filaments, except for the 40  
 276 wt.-% and the 0.84 mm extrusion nozzle diameter.

277

278 For the 40 wt.-% ink filament extruded with a 0.84 mm extrusion nozzle, all avail-  
 279 able experimental data have been used. It was found, after trials and errors, that setting  
 280  $\alpha_j = 4$  for  $t \in [0, 2.5]$  seconds for the tension-relaxation test (and  $\alpha_j = 1$  otherwise) led  
 281 to model predictions fitting relatively well all the experimental data on Figures 7 and 8.

282

283 All these results show that a linearly viscoelastic constitutive theory might be a  
 284 practical choice for modelling the mechanical behaviour of such ink filaments. Even  
 285 though some discrepancies can be observed for the 40 wt.-% filament extruded with a  
 286 0.84 nozzle, the authors believe that the models obtained in this study could be useful  
 287 for predicting scaffold behaviour and contributing to the optimization of DWM struc-  
 288 tures.

289

290 Subjecting the various filaments to different load histories (as was done for the  
 291 40 wt.-% filament extruded with a 0.84 nozzle) would allow determining the degree

292 of precision that can be expected from linearly viscoelastic constitutive theories for  
293 modelling the mechanical behaviour of ink filaments. The tension testing machine  
294 developed in this study is capable of generating a wide spectrum of uni-dimensionnal  
295 load histories and could be used to this end.

#### 296 4. Conclusions

297 A custom-designed tensile machine has been used to characterize organic ink based on a  
298 mixture of petroleum jelly and a specific amount of microcrystalline wax when extruded  
299 and deposited as filaments. It has been shown that the ink filament response presents a  
300 strain rate dependence. We observed that increasing microcrystalline wax amount and  
301 decreasing the inner diameter nozzle lead to a stiffer ink filament. A tension-relaxation  
302 test conducted on a 40 wt.-% ink filament has shown that the material exhibits a  
303 viscoelastic behaviour. Constitutive theory parameters identification demonstrated that  
304 ink filaments behaviour is dominated by a delayed response and prediction responses  
305 based on these parameters are close to the experimental data. Further refinements  
306 are needed to improve the force sensor precision in order to maximize the duration of  
307 the tensile and tension-relaxation tests according to the ASTM C1557-03<sup>E1</sup> Standard.  
308 These improvements will provide important information on the mechanical behaviour of  
309 ink filaments and their constitutive theory parameters under various loads which may  
310 find interest in being implemented in finite elements programs to simulate not only ink  
311 filaments in suspension but also scaffolds. We foresee that a similar approach could be  
312 applied for other ink materials such polyelectrolytes and nanocomposites.

#### 313 Acknowledgments

314 The authors wish to acknowledge financial support from Natural Sciences and  
315 Engineering Research Council of Canada (NSERC).

#### 316 References

- 317 [1] N. Bao, J. Wang, and C. Lu. Recent advances in electric analysis of cells in microfluidic systems.  
318 *Journal of Analytical and Bioanalytical Chemistry*, 391(3):933 – 942, 2008.
- 319 [2] T. A. Franke and A. Wixforth. Microfluidics for miniaturized laboratories on a chip.  
320 *ChemPhysChem*, 9(15):2140 – 2156, 2008.
- 321 [3] A. Balasubramanian, B. Bhuvu, R. Mernaugh, and F. R. Haselton. Si-based sensor for virus  
322 detection. *Sensors Journal*, 5(3):340 – 344, 2005.
- 323 [4] S. Bangalore Prakash and P. Abshire. Tracking cancer cell proliferation on a cmos capacitance  
324 sensor chip. *Biosensors & Bioelectronics*, 23(10):1449 – 1457, 2008.
- 325 [5] J. A. Lewis. Direct ink writing of 3d functional materials. *Advanced functional materials*,  
326 16(17):2193 – 2204, 2006.
- 327 [6] R. Narayan. Recent developments in rapid prototyping of biomaterials. *Biotechnology Journal*,  
328 2(11):1340 – 1341, 2007.
- 329 [7] D. Therriault, S. White, and J. Lewis. Chaotic mixing in three-dimensional microvascular networks  
330 fabricated by direct-write assembly. *Nature Materials*, 2:265 – 271, 2003.

- 331 [8] M. Kuhn, T. Napporn, M. Meunier, S. Vengallatore, and D. Therriault. Direct-write  
332 microfabrication and electrochemical testing of single-chamber micro solid oxide fuel cells.  
333 *Journal of Micromechanics and Microengineering*, 18, 2008.
- 334 [9] R. Bey-Oueslati, D. Therriault, and S. Martel. Pcb-integrated heat exchangers for cooling  
335 electronics using microchannels fabricated with the direct-write method. *IEEE Transaction*  
336 *on Components and Packaging Technologies*, 31(4):869 – 874, 2008.
- 337 [10] D. Therriault, R. Shepherd, S. White, and J. Lewis. Fugitive inks for direct-write assembly of  
338 three-dimensional microvascular networks. *Advanced Materials*, 17:395 – 399, 2005.
- 339 [11] D. Therriault, S. White, and J. Lewis. Rheological and structural behavior of fugitive inks for  
340 direct-write assembly. *Applied Rheology*, 17(1):10112–1 – 10112–8, 2007.
- 341 [12] J. Bruneaux, D. Therriault, and M.-C. Heuzey. Micro-extrusion of organic ink for direct-write  
342 assembly. *Journal of Microengineering and Micromechanics*, 18:115020–1 – 115020–11, 2008.
- 343 [13] J. E. Smay, J. Cesarano III, and J. A. Lewis. Colloidal inks for directed assembly of 3-d periodic  
344 structures. *Langmuir*, 18(14):5429 – 5437, 2002.
- 345 [14] M. Lévesque, K. Derrien, D. Baptiste, and M. Gilchrist. On the development and parameter  
346 identification of schapery type constitutive theories. *Mechanics of Time-Dependent Materials*,  
347 12(2):95 – 127, 2008.
- 348 [15] M. Lévesque, M. D. Gilchrist, N. Bouleau, K. Derrien, and D. Baptiste. On applying the  
349 numerical laplace-carson transform inversion to the homogenisation of randomly reinforced linear  
350 viscoelastic media. *Computational Mechanics*, 40(4):771 – 789, 2007.
- 351 [16] R.B. Barello and M. Lévesque. Comparison between the relaxation spectra obtained from  
352 homogenization models and finite elements simulation for the same composite. *International*  
353 *Journal of Solids and Structures*, 45:850 – 867, 2008.
- 354 [17] H. J. Lee, H. S. Choi, C. S. Han, N. K. Lee, G. A. Lee, and T. H. Choi. A precision alignment  
355 method of the micro tensile testing specimen using mechanical gripper. *Journal of Materials*  
356 *Processing Technology*, 187 - 188:241 – 244, 2007.