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1 **Instability and freezing in a solidifying melt conduit**

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6 October 23, 2010

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8 Previous works have shown that when liquid flows in a pipe whose boundary
9 temperature is below freezing, a tubular drainage conduit forms surrounded by solidified
10 material that freezes shut under the appropriate combination of forcing conditions. We
11 conduct laboratory experiments with wax in which the tube freezes shut below a certain
12 value of flux from a pump. As the flux is gradually decreased to this value, the total
13 pressure drop across the length of the tube first decreases to a minimum value and then
14 rises before freezing. Previous theoretical models of a tube driven by a constant pressure
15 drop suggest that once the pressure minimum is reached, the states for a lower flux
16 should be unstable and the tube should therefore freeze up. In our experiments, flux and
17 pressure drop were coupled, and this motivates us to extend the theory for low-Reynolds
18 number flow through a tube with solidification to incorporate a simple pressure drop-flux
19 relationship. Our model predicts a steady-state relationship between flux and pressure
20 drop that has a minimum of the pressure as the flux is varied. The stability properties of
21 these steady states depend on the boundary conditions: for a fixed flux, they are all stable,
22 whereas for fixed pressure drop, only those with a flux larger than that at the pressure

23 drop minimum are stable. For a mixed pressure-flux condition, the stability threshold of
24 the steady states lies between these two end members. This provides a possible
25 mechanism for the experimental observations.

26 **1. Introduction**

27 Injected liquids that freeze as they flow are common in many areas of engineering
28 (injection molding, freezing, metallurgy) as well as in earth and planetary sciences (lava
29 tubes, magma conduits, glaciology, and magma fissure flows). In such cases, liquid flows
30 through a region whose boundary temperature is below the solidification temperature of
31 the liquid, so that advection of heat by the warm liquid acts in tandem with removal of
32 heat by the boundary. In some cases, the cooling is weak enough that solid may form at
33 the boundary but leave a central melted tube where liquid flows. In other cases the entire
34 body of liquid may freeze so that all flow ceases. It is useful to know the conditions that
35 are necessary for such freezing.

36 In the geophysical literature, the pioneering study of the dynamics of melting and
37 solidifying material was for flow up a fissure with variable gap width [*Bruce and*
38 *Huppert*, 1989, 1990], where conditions for melt-back (widening) or solidification
39 (narrowing) of the gap are calculated from thermal energy budgets. This was followed by
40 many studies of the dynamics of either fissure flow or lava dynamics, investigating
41 situations such as the temperature distribution and velocity profile in a magma tube, or
42 the driving pressure required to keep it open [e.g. *Sakimoto and Zuber*, 1998, *Dragoni et*
43 *al.*, 2002, *Sakimoto and Gregg*, 2001, *Klingelhofer et al.*, 1999]. These studies invariably
44 use simplified, time-independent geometries for the tube boundary, and generally, little
45 analysis has been made of the stability of the flows. A notable exception is the theoretical

46 study by Lister and Dellar, [1996], in which the cooling occurs at infinity and therefore
47 no steady-state tube is possible.

48 For engineering purposes, numerous studies focus on flow of a liquid in a
49 container whose walls are below the freezing temperature. Applications include injection
50 molding, the freezing of water, the condensation of water vapor in ducts, and metal
51 casting, among others. For example, experiments with water demonstrate the focusing of
52 flow into a narrow region along with the formation of waves of solid on the walls, and in
53 some cases freeze-up [Zerkle and Sunderland, 1968 Mulligan and Jones, 1976 Hirata
54 and Ishihara 1985, Weigand et al., 1997]. A common feature is that the curve of steady-
55 state pressure drop against flux exhibits sizeable curvature, in many cases reaching a
56 minimum such that as the flux is gradually decreased, the pressure drop first decreases,
57 then increases, a result that has been recovered in theoretical studies [Zerkle and
58 Sunderland, 1968, Lee and Zerkle, 1969.] If the flow is driven by imposing a fixed
59 pressure drop, however, the low-flux branch of this curve, where pressure drop increases
60 with decreasing flux, is unstable: a perturbation making a smaller cross sectional area
61 produces more drag, which produces slower flow that leads to colder liquid and more
62 solidification and finally to total freezing [Sampson and Gibson, 1981, Richardson 1985].
63 If, instead, the flow is driven by a pump imposing a fixed flux, the steady state is
64 presumed to be stable, although a complete stability analysis has never been done; a
65 smaller cross sectional area makes a faster flow that brings warmer fluid from upstream
66 to the region, which widens the perturbation. The constant flux upstream condition is
67 widely used in theories that calculate the solid accumulation along flow ducts of assorted
68 material properties and shapes [e.g., Mulligan and Jones, 1976, Epstein and Chueng,

69 1983, *Richardson*, 1986], but such problems do not exhibit flow freeze-up from an
70 instability.

71 Since theory shows that stability depends on the particular type of flow boundary
72 condition that is imposed at the upstream end, our attention here is focused upon the
73 stability of solidifying flow with a more general upstream condition than either constant
74 flux or constant pressure drop. We tackle the question of stability with both experiment
75 and theory. First, we describe laboratory experiments of flow through a pipe whose
76 temperature is held below the solidus, in which there was a coupling between flux and
77 pressure drop (Section 2). The flow froze when the steady-state flux was below a certain
78 value. As the steady-state flux was decreased in successive experiments to this value, the
79 pressure drop across the tube reached a minimum and then increased before freeze-up.
80 This result is not explained by either constant flux or constant pressure drop models, one
81 of which suggests freeze-up should never occur, and the other that it should occur as soon
82 as the pressure minimum is reached. It motivates us to investigate the stability of low-
83 Reynolds number flow through a tube using a standard idealized theoretical model with
84 the addition of a mixed pressure-flux upstream driving condition (Section 3). Essentially,
85 we suppose the tube drains from an upstream reservoir into which fluid is pumped at a
86 constant rate, so the total amount of fluid in the reservoir determines the driving pressure
87 and therefore the flux through the tube. Naturally, this new upstream condition is
88 intended to be a more realistic model both of conditions in our experiment as well as in
89 some types of geological melt conduits, and possibly in some engineering applications. A
90 linear stability analysis shows that the mixed upstream condition allows the stable range
91 of flow to extend to lower values of flux that are unstable for fixed pressure drop. Thus, it

92 is in qualitative accord with the laboratory results. In addition, the theory predicts an
93 oscillatory instability that has not been found in previous theoretical studies. Numerical
94 simulations recover both new features (Section 3.4). In Section 3.5 we show how the
95 basic model (without the stability results) can be used straightforwardly to provide a
96 realistic constraint on the length of geological melt conduits.

97 The central implication of these results is that stability is very sensitive to the
98 upstream conditions that drive the melt through the tube. This sensitivity may be one
99 mechanism behind the complex nature of many real solidifying flows in nature and
100 industry.

101 2. *Experiments with freezing of flow through a tube.*

102

103 We performed experiments with flow through a chilled circular pipe, whose setup
104 is shown in Figure 1. The pipe was a standard glass condenser for a chemistry laboratory
105 with a central glass pipe of radius $r_0 = 0.49 \times 10^{-3}$ m surrounded by a sleeve (see Table 1
106 for list of symbols). The length of the portion of the pipe surrounded by this sleeve was
107 $L = 0.18$ m. The sleeve was flushed by water from a constant temperature bath at
108 temperature T_0 that was accurate to ± 0.1 °C. The central axis of the condenser was
109 placed horizontally. Liquid at 20 °C was fed from a constant displacement metered
110 pump into one end of the condenser. The pump volume flux rate (henceforth simply
111 called either flux or, in case of a pump setting, the pumping rate) was calibrated to $\pm 2\%$.
112 The other end was the tube exit fitted with a rubber stopper with a flat notch cut along the
113 top. The liquid exited the glass tube by flowing over this notch; therefore, the stopper

114 served as a miniature dam so that the pipe within the condenser remained filled with
115 liquid at all times with no air traveling upstream from the exit into the tube. A photograph
116 of the outlet with the stopper removed after a run shows a circular drainage channel
117 surrounded by solid (Figure 2). The ridges in the solid are evidence of uneven
118 solidification whose origin will not be studied further here. The liquid was 1-Octadecene
119 (Chevron Phillips C18, kindly donated),. In this study, we simply call this material a
120 wax. The freezing point (solidus temperature) is $T_s = 17.8^\circ\text{C}$ and the pour point is half a
121 degree higher at 18.3°C , indicating that viscosity increases greatly close to the solidus.
122 The specifications for the liquid state are: a thermal conductivity of $k = 0.114\text{ W/m}^\circ\text{K}$, a
123 specific heat of $c_p = 2.26 \times 10^3\text{ J/kg}^\circ\text{K}$ with significant changes in value near freezing
124 temperature (Bundhu et al. 1998), a density of $\rho = 785\text{ kg/m}^3$ (these three give a thermal
125 diffusivity of $\kappa = 0.64 \times 10^{-7}\text{ m}^2\text{ s}^{-1}$) and kinematic viscosity values of $\nu = 8.28 \times 10^{-6}$
126 $\text{m}^2\text{ s}^{-1}$ at 31°C and $\nu = 3.8 \times 10^{-6}\text{ m}^2\text{ s}^{-1}$ at 37.8°C . Also, the fluid is very hygroscopic.
127 Since the model developed in subsequent sections assumes constant material properties,
128 the fact that viscosity and specific heat changes greatly in the temperature range of
129 interest means that we will only be able to compare the experimental results with
130 prediction qualitatively.

131 For all experiments, the temperature of the liquid pumped into the condenser was
132 $T_i = 20^\circ\text{C}$. After starting the liquid pump, the temperature of the water flushing the
133 sleeve was set to a value below the solidus so that the wax became solid along the inner
134 radius of the glass pipe as sketched in Figure 1, with flow occurring in a central liquid
135 tube. The liquid tube radius varied in the flow direction and it was a function of the
136 pumping rate and sleeve temperature. We measured pressure immediately upstream of

137 the condenser by splitting the upstream plastic tubing with a Y connection. The tube in
138 one side of the Y was the input to the condenser and the other plastic tube was held
139 vertically next to a centimeter scale to allow a measurement of pressure of the upstream
140 fluid. Since pressure at the downstream end was fixed at atmospheric pressure, the
141 elevation of the liquid surface in the vertical plastic tube above the elevation of the outlet
142 was proportional to pressure drop across the condenser. This elevation was read to a
143 precision of 1 mm. The vertical tube is also a storage region for liquid supplied by the
144 pump. In fact, the difference between the flux of the pump and the flux out through the
145 condenser is proportional to the rate of change of height in the vertical pressure tube.
146 This provides a mixed pressure-flux upstream boundary condition to the flow through the
147 condenser. The exact expression for this will be derived in the next section.

148 The top of the vertical plastic tube was bent over and extended back to the wax
149 reservoir as an overflow. If upstream pressure became too great, the overflowing liquid
150 indicated freeze-up of the tube

151 The procedure for these experiments at the beginning of each day was to start
152 with everything at room temperature so the wax was completely liquid. A run
153 commenced by turning on the wax pump to a desired pumping rate and then changing the
154 cold bath temperature from 20 °C to the desired value, which we call T_0 . After about 15
155 minutes, the wax solidified along the inner radius of the tube and the flow continued
156 through the liquid tube. The elevation in the vertical tube was measured many times until
157 the value was steady, and then the final value of pressure (in units of vertical elevation)
158 was recorded. The flux was also measured then.

159 Figure 3 shows the elevation of the liquid surface in the vertical tube versus the
160 imposed pumping rate, or flux for many runs in experiments with cold bath temperatures
161 set to two different values: $T_0 = 5.0$ °C and $T_0 = 10.0$ °C. At both temperatures the
162 flowing liquid froze shut at a pumping rate approximately 5% below the measurement on
163 the extreme left. To the right of the freezing point, the inverse relation between the
164 pressure and pumping rate was unmistakable. For $T_0 = 10.0$ °C, pressure increased
165 slightly with pumping rate for flux $Q_i > 0.5 \times 10^{-6}$ m³ s⁻¹ but for $T_0 = 5.0$ °C, a pressure
166 increase with flux is not visible. The errors for the pressure measurement and for the
167 calibration of the pumping rate are approximately the size of the symbols. Since
168 obviously the scatter about a smooth curve for all the data is considerable, we concluded
169 after careful checking that the scatter is not from errors in measurement. In addition, we
170 conducted long runs to determine whether the scatter was due to the experiment duration
171 being too short. For all these experiments (which were conducted for more than two
172 hours each, and comprise 70% of the data points), such scatter persisted even though
173 the pressure reading had been constant for the entire second hour. Therefore, we believe
174 the scatter is a basic feature and the scatter might possibly be due to small differences in
175 the detailed shape of each frozen solid. In support of this, Figure 2 shows irregularities in
176 the solid surface near the exit.

177 The experiment results are scaled by noting that the experimental flow tube has
178 the following variables: the glass tube radius r_0 , tube length L , fluid viscosity μ , fluid
179 density ρ , fluid thermal diffusivity κ , temperature at the inlet T_i , temperature of the
180 surface of the tube T_0 , temperature of solidus T_s , and flux of the liquid initially entering
181 upstream Q_i . This totals 9 variables with four units: temperature, force, length and time.

182 Therefore, five dimensionless numbers are needed. Two of them are simply temperature

183 ratios, but they are best combined and expressed as $T_n = \frac{T_s - T_0}{T_i - T_s}$. A third is aspect ratio

184 of the tube r_0/L . A fourth is Prandtl number $Pr = \nu/\kappa$, and the last is nondimensional

185 flux $q_i = \frac{2}{\kappa\pi L} Q_i$. In addition, we calculate a value of nondimensional pressure drop

186 $\Delta P = \frac{r_0^4}{4\mu\kappa L^2} P$, where P is the pressure above atmosphere pressure at the upstream end

187 .

188 Using the values for this liquid, the Prandtl number is $Pr = 129$. Using the tube

189 length and radius, and using the magnitude for flux near the minimum of about

190 $Q_i = 0.3 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$ from Figure 3, we get $q_i = 15$. The magnitude of scaled pressure

191 from the same figure is found using the hydrostatic equation for pressure $P = \rho g H$,

192 where acceleration from gravity is g and a typical elevation of wax in the vertical

193 pressure measuring tube is $H = 0.02 \text{ m}$. From this, we get $\Delta P = 1650$.

194 Next, the values of actual critical fluxs for freezing were checked by four precise

195 experiments at four different values of $T_0 = 2.5, 5.0, 7.5,$ and $10.0 \text{ }^\circ\text{C}$. For each of these

196 values, an experimental run started with the pump set at a value that allowed continuous

197 flow. Then, the freezing point was approached by decreasing the pumping rate by 5%

198 increments and waiting an hour or more to see if the flow froze. If the flow did not freeze

199 after that time interval, another decrease was made. The aggregate time for each run was

200 many hours. The lowest values of pumping rate at the above four temperature settings are

201 $0.42, 0.23, 0.18,$ and $0.16 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$, successively, These correspond to non-

202 dimensional values of $q_i = 23.2, 12.7, 9.95,$ and 8.84 at $T_n = 6.95, 5.82, 4.68,$ and $3.55,$

203 respectively. Flow ceased by freezing shut for incrementally changed pumping rates that
204 were approximately 5% below these rates.

205 In experiments using more than the 5% incremental decrease in pumping rate
206 from one experiment to the next, the critical flux for freezing was measurably larger. For
207 example, the wax always froze shut for experiments at $T_0 = 10^{\circ}\text{C}$ with a steady pumping
208 rate of $0.36 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$ and then after steady flow developed were given a 33% decrease
209 in pumping rate to $0.24 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$ ($q_i=19.89$ to 13.26), The exact reason why a large
210 incremental decrease leads to a higher critical flux than the value with a 5% incremental
211 decrease, which in this case is $0.16 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$ ($q_i = 8.84$), is unknown. Possibly the
212 upstream pressure cannot build up rapidly enough to allow sufficient flux through the
213 melt region when the interior radius shrinks.

214 After a steady flow developed, the stopper at the exit was removed to view the
215 inner conduit radius by looking into the end of the pipe. A light beam from a slide
216 projector at a right angle to the tube and directed at the end of the tube far from the
217 camera illuminated fluid upstream as the white circle in Figure 2. Regrettably, we are
218 skeptical of using such images to attempt to measure the diameter of the liquid conduit.
219 Clearly, there was large distortion of the light as it passed to the camera across the curved
220 liquid/air surface. Also, each light beam arriving to the camera from the inside of the
221 liquid tube was bent by the axial temperature distribution within the liquid tube with the
222 axial equivalent of the mirage effect. Therefore, no optical measurements of the tube
223 radius as a function of flow rate and sleeve temperature were attempted.

224 If the flux and the bath temperature were slightly above the values that gave
225 freezing, the flow was easily made to freeze even with very small disturbances. For

226 example, with a sleeve temperature of $2.5, ^\circ\text{C}$ and pumping rate of $0.42 \times 10^{-6} \text{ mm}^3 \text{ s}^{-1}$,
227 when the pump was stopped for five seconds, the flow ceased and never started again.
228 Conversely, with the same initial conditions the flow resumed most of the time if the
229 pump was stopped for three seconds, and it always resumed if the pump was stopped for
230 only one second. We also found that a piece of very fine copper wire inserted into the
231 liquid hole readily nucleated a freezing event.

232 3. *Flow through a tube, theory*

233 3.1 *Fundamental Equations*

234 We begin the analysis by reviewing a standard theoretical model for a melt conduit of
235 flow at low Reynolds number into a long cold pipe [eg *Zerkle and Sunderland*, 1968].
236 The pipe has a fixed length L in the x -direction and it has a perfectly circular cross-
237 section with constant radius r_0 (Figure 4). Liquid enters the pipe at a uniform initial hot
238 temperature T_i and it flows with laminar flow. The boundary of the pipe is maintained at
239 a constant temperature T_0 that is colder than the solidification temperature T_s . The
240 temperature varies continuously from $T = T(\theta, x, t) > T_s$ in the liquid at the center of the
241 tube, to T_0 at r_0 . Solid material forms a tube of radius $a(x, t)$ at the isotherm $T = T_s$.

242 A number of assumptions are made to make the model analytically tractable. A full
243 list can be found in *Zerkle and Sunderland* [1968], but we mention those that will be
244 most important. First, the basic flow is made as simple as possible by assuming that there
245 are constant material properties, a simple cutoff solidification temperature, and no
246 buoyancy force. Second, the Reynolds number is small enough for there to be no
247 turbulence and no inertia in the momentum equation. Third, the length L is assumed to be
248 large enough compared to r_0 that changes in the along-tube direction x are slow. Finally,

249 the Stefan number is assumed to be large, so that the solidification process and
 250 corresponding motion of the crust are much slower than the thermal, advective, or
 251 viscous timescales. Therefore time-derivatives are only retained in the equation for the
 252 radius, and while other fluid variables are time-dependent, they are only quasi-steadily so
 253 via their dependence on the radius. We now proceed to introduce the basic equations.

254 The velocity in the downstream direction is given by the well-known equation for

255 flow at low Reynolds number [eg *Turcotte and Schubert, 2002*], $\frac{\partial P}{\partial x} = \mu \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u}{\partial r} \right)$,

256 where $\partial P / \partial x$ is pressure gradient in the axial direction, u is velocity in the axial direction,

257 μ is fluid viscosity and r is the radial coordinate. The radial velocity v can be found

258 from the condition of non-divergence, and is non-zero because the radius of the tube

259 changes in the flow direction. The solution for u with the boundary condition $u=0$ at

260 $r=a(x,t)$ is Poiseuille flow $u = -\frac{\partial P}{\partial x} \frac{(a^2 - r^2)}{4\mu}$. Integrating over the area determines the

261 flux Q whose relation to the pressure gradient is

262
$$\frac{\partial P}{\partial x} = -\frac{8\mu Q}{\pi a^4}, \quad (3.1)$$

263 so the velocity can also be written as

264
$$u = \frac{2Q}{\pi a^2} \left(1 - (r/a)^2 \right). \quad (3.2)$$

265 In the solid, the temperature field T_e satisfies a diffusion equation when the x -

266 derivatives and time-derivatives are neglected:

267
$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T_e}{\partial r} \right) = 0 \quad , \quad (3.3)$$

268 with the boundary conditions $T_e|_{r=r_0} = T_0$, $T_e|_{r=a} = T_s$. This can be solved to give

269
$$T_e = \frac{T_0 - T_s}{\ln \frac{r_0}{a}} \ln \frac{r}{a} + T_s \quad . \quad (3.4)$$

270 In the liquid, the temperature field is determined by a balance between advection

271 and diffusion when time-derivatives are neglected:

272
$$u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial r} = \kappa \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) \quad , \quad (3.5)$$

273 with boundary conditions $T(r = a) = T_s$, $T(x = 0) = T_i$, $\frac{\partial T}{\partial r} \Big|_{r=0} = 0$.

274 It is more convenient to solve this by defining a new variable $\eta = r / a$, which scales the

275 radial coordinate by the radius of the tube, so that streamlines of the flow are lines of

276 constant η . Under this transformation equation (3.5) becomes

277

278
$$\frac{2Q}{\kappa \pi a^2} (1 - \eta^2) \frac{\partial T}{\partial x} = \frac{1}{a^2} \frac{1}{\eta} \frac{\partial}{\partial \eta} \left(\eta \frac{\partial T}{\partial \eta} \right) \quad (3.6)$$

279

280 with boundary conditions $T|_{\eta=1} = T_s$, $\frac{\partial T}{\partial \eta} \Big|_{\eta=0} = 0$, $T|_{x=0} = T_i$.

281 The final equation is for the radius. The time-dependent equation for the radius is
 282 a standard Stefan equation [e.g. *Turcotte and Schubert, 2002*]

283

$$284 \quad \frac{L_H}{c_p} \frac{\partial a}{\partial t} = \kappa \left(\left. \frac{\partial T_e}{\partial r} \right|_{r=a} - \left. \frac{\partial T}{\partial r} \right|_{r=a} \right), \quad (3.7)$$

285

286 where κ is thermal diffusivity of both the liquid and solid, which are assumed here to be
 287 equal in magnitude, L_H is the latent heat of solidification, and c_p is the heat capacity of
 288 the liquid. The rate of change of the radius of the tube is proportional to the difference in
 289 heat flux at the boundary of the tube, which, by the slowly-varying-in- x assumption, is
 290 the flux in the radial direction only.

291

292 **3.2 Steady-state Solutions**

293 We first consider the solution for the steady-state of the model, given by the
 294 steady components of (3.1, 3.3, 3.6, 3.7) with the corresponding boundary conditions.

295 The equations are non-dimensionalized with $x = L\chi$, $a = r_0\alpha$, $\frac{T - T_s}{T_i - T_s} = \theta$,

$$296 \quad \frac{T_e - T_s}{T_i - T_s} = \theta_e, \quad Q = \frac{\kappa L \pi}{2} q, \quad P = \frac{4\mu\kappa L^2}{r_0^4} p, \quad \text{and} \quad u = \frac{\kappa L}{r_0^2} u'. \quad \text{Pressure is non-}$$

297 dimensionalized so it remains in the balance to first order, and flux is non-

298 dimensionalized so that the effect of conductive cooling is balanced by advection. The

299 model depends on a dimensionless imposed temperature difference

300
$$T_n = \frac{T_s - T_0}{T_i - T_s} . \quad (3.8)$$

301 The non-dimensional velocity and the temperature in the solid are

302
$$u' = \frac{q}{\alpha^2} (1 - \eta^2) \quad , \quad \theta_e = \frac{T_n \ln \eta}{\ln \alpha} \quad (\eta \geq 1) \quad (3.9)$$

303

304 and the pressure drop across the tube Δp is related to the flux by:

305

306
$$\Delta p = q \int_0^1 \frac{1}{\alpha^4} d\chi . \quad (3.10)$$

307

308 The steady non-dimensional internal temperature equation is

309
$$q(1 - \eta^2) \frac{\partial \theta}{\partial \chi} = \frac{1}{\eta} \frac{\partial}{\partial \eta} \left(\eta \frac{\partial \theta}{\partial \eta} \right) . \quad (3.11)$$

310

311 This can be solved by separation of variables to give

312
$$\theta(\chi, \eta) = \sum_n A_n e^{-\lambda_n^2 \chi / q} \phi_n(\eta) , \quad (3.12)$$

313 where λ_n, ϕ_n are the eigenvalues and eigenfunctions of the problem

314
$$\frac{1}{\eta} \frac{\partial}{\partial \eta} \left(\eta \frac{\partial \phi_n}{\partial \eta} \right) + \lambda_n^2 (1 - \eta^2) \phi_n = 0 , \quad \phi_n(0) = 1, \phi_n(1) = 0, \phi'_n(0) = 0 .$$
 The solution was

315 originally found by *Graetz* [1883] for flow of uniform viscosity through a pipe of

316 constant radius, and was modified for steady flow with solidification as in this

317 configuration by *Zerkle and Sunderland* [1968]. The A_n are constants determined from

318 the upstream temperature distribution. A more complete discussion of this solution,
 319 including numerical values, is given in the appendix of *Sakimoto and Zuber* [1998]. In
 320 steady-state, the dimensionless equation at the liquid solid interface becomes

321

$$322 \quad \left. \frac{\partial \theta}{\partial \eta} \right|_{\eta=1} = \left. \frac{\partial \theta_e}{\partial \eta} \right|_{\eta=1} . \quad (3.13)$$

323

324 Using (3.9) and (3.12), we calculate

$$325 \quad \left. \frac{\partial \theta_e}{\partial \eta} \right|_{\eta=1} = \frac{T_n}{\ln \alpha} ,$$

326

$$327 \quad \left. \frac{\partial \theta}{\partial \eta} \right|_{\eta=1} = \sum G_n e^{-\lambda_n^2 \chi / q} , \quad \text{where} \quad G_n = A_n \left. \frac{\partial \phi_n}{\partial \eta} \right|_{\eta=1} ,$$

328

329 so the radius of a steady-state tube is

$$330 \quad \alpha(\chi) = \exp \left(\frac{T_n}{\sum G_n e^{-\lambda_n^2 \chi / q}} \right) . \quad (3.14)$$

331

332 Profiles of α for several different values of q are shown in Figure 5a. Note the
 333 relation between α , q , and Δp . If flux q is prescribed then (3.14) gives an explicit
 334 solution for α , while if Δp is prescribed it must be solved in conjunction with (3.10),
 335 which provides a transcendental integro-differential equation for α . Figure 6a shows the

336 pressure drop as a function of flux for a steady-state tube, for a particular choice of
337 temperature constant. This has a minimum Δp_c at a critical flux q_c , suggesting that when
338 $\Delta p > \Delta p_c$ there are two solutions for a steady-state tube and when $\Delta p < \Delta p_c$ there are no
339 possible tubes, a fact which has been verified analytically in [Holmes, 2007]. The critical
340 pressure drop $\Delta p_c(T_n)$ and critical flux at which it is attained $q_c(T_n)$ are shown in Figure
341 5b.

342 Note the qualitative similarities between the analytic pressure drop-flux
343 relationship in Figure 6a and the experimental results in Figure 3: as flux is decreased
344 there is a very weak decline in pressure drop, and then a sudden sharp increase for low
345 values of flux.

346

347 ○ **3.3. Linear stability analysis**

348 To investigate stability we introduce an upstream condition with an additional
349 parameter to capture each of three possibilities: (i) constant flux, (ii) constant pressure,
350 and (iii) a model allowing the two variables to co-vary. One assumes that the tube is fed
351 from an upstream reservoir that in turn is fed by a steady volume flux of rate Q_i . (The
352 model can also be derived by assuming the upstream reservoir is elastic.) Flow from the
353 reservoir obeys the equation

354

355
$$A \frac{dH}{dt} = Q_i - Q ,$$

356

357 where A is the cross-sectional area of the reservoir and H is fluid elevation in it. The
 358 downstream end of the tube is open and hence at atmospheric pressure, so the pressure
 359 drop across the tube is given by

$$360 \quad \Delta P = g\rho H.$$

361

362 Letting the timescale be Sr_0^2/κ , where Stefan number is $S = L_H/c_p(T_i - T_s)$, and
 363 non-dimensionalizing the other scales as before, leads to the non-dimensional system

364

$$365 \quad \frac{\partial \alpha}{\partial t} = \frac{1}{\alpha} (E(\alpha) - I(\chi, q)) \quad (3.15a)$$

$$366 \quad \frac{d\Delta p}{dt} = \tau(q_i - q) \quad (3.15b)$$

$$367 \quad \Delta p = q \int_0^1 \frac{1}{\alpha^4} d\chi \quad (3.15c)$$

368 where the temperature gradient in the solid at the solid-liquid interface is

$$369 \quad E(\alpha) = \left. \frac{\partial \theta_e}{\partial \eta} \right|_{\eta=1} = \frac{T_n}{\ln \alpha}, \text{ and the temperature gradient in the liquid at the interface is}$$

$$370 \quad I(\chi, q) = \left. \frac{\partial \theta}{\partial \eta} \right|_{\eta=1} = \sum G_n e^{-\lambda_n^2 \chi / q}.$$

371 This model has a new non-dimensional parameter $\tau = \frac{\pi g S r_0^6}{8 A \nu \kappa L}$, which measures the rate

372 of change of the upstream pressure relative to the rate of change of the radius of the

373 interface, and is proportional to the Stefan number times a thermal response time r_0^2/κ

374 divided by the hydraulic reservoir response time ALv/gr_0^4 . The latter is the exponential
 375 time for a viscous fluid to empty the reservoir with no solidification ($T_n \rightarrow 0$).

376 The model also depends on the non-dimensional flux q_i into the upstream
 377 reservoir. Therefore, the dynamics of (3.15) are determined by the three parameters
 378 T_n, τ, q_i . When $\tau \ll 1$, the elevation, or pressure in the reservoir adjusts extremely slowly
 379 to changes in the flux, and by extension the radius of the tube, so the system should
 380 behave as if the pressure drop were held constant, with a constant-pressure-drop system
 381 recovered exactly when $\tau = 0$. When $\tau \gg 1$, the pressure in the reservoir adjusts rapidly
 382 to the flux into the reservoir so the system should behave as if the flux through the tube
 383 were held constant. Thus, setting different values of τ allows us to quantitatively
 384 interpolate between constant flux and constant pressure drop conditions.

385 Let us now examine the linear stability of (3.15). Expanding to first order in small
 386 ε , $q = q_0 + \varepsilon q_1$, $\alpha = \alpha_0 + \varepsilon \alpha_1$, and $\Delta p = p_0 + \varepsilon p_1$ (note we have dropped the Δ symbol
 387 for the pressure drop steady-states and perturbations), the steady-state is

388
 389
$$q_0 = q_i \tag{3.16a}$$

390
$$\alpha_0 = \alpha_0(\chi, q_i) = \exp\left(\frac{T_n}{\sum G_n e^{-\lambda_n^2 \chi / q_i}}\right) \tag{3.16b}$$

391
$$p_0 = q_i \int_0^1 \frac{1}{\alpha_0^4} d\chi \tag{3.16c}$$

392

393 and the $O(\varepsilon)$ parts are

394

$$395 \quad \frac{\partial \alpha_1}{\partial t} = \frac{1}{\alpha_0} \left(\frac{dE}{d\alpha} \Big|_{\alpha_0} \alpha_1 - \frac{\partial I}{\partial q} \Big|_{q_0} q_1 \right) \quad (3.17a)$$

$$396 \quad \frac{dp_1}{dt} = -\tau q_1 \quad (3.17b)$$

$$397 \quad q_1 = \frac{p_1 - \frac{\delta \Delta p}{\delta \alpha} \Big|_{\alpha_0, q_0} [\alpha_1]}{\frac{\partial \Delta p}{\partial q} \Big|_{\alpha_0, q_0}} \quad (3.17c)$$

398

399 The forms of some of the functions are given in Table 2. In these equations we
 400 have taken care to distinguish between partial derivatives and functional derivatives, by
 401 using the symbol ∂ for a partial derivative and δ for a functional derivative, which
 402 results in a linear operator. We simplify notation by writing $E_\alpha(\chi) \equiv \frac{1}{\alpha_0} \frac{dE}{d\alpha} \Big|_{\alpha_0}$. Let us
 403 analyze the three different boundary conditions in turn.

404 **Case (i): constant flux.** The stability of the constant flux case is simple to
 405 analyze separately. Replacing (3.17b) with the condition $q_1 = 0$ and substituting for
 406 $E(\alpha)$, equation (3.17a) becomes

407

$$408 \quad \frac{d\alpha_1}{dt} = E_\alpha \alpha_1 = \frac{-T_n \alpha_1}{\alpha_0^2 \ln^2 \alpha_0}.$$

409

410 Since both $\alpha_0, T_n > 0$, we have that $\text{sgn}(d\alpha_1/dt) = -\text{sgn}(\alpha_1)$ for every χ , so this
 411 equation is sign-definite and hence linearly stable.

412

413 **Case (ii): constant pressure.** This case was first analyzed by *Sampson and*
 414 *Gibson*, [1981]. Recall that for a given pressure drop there are two possible steady-state
 415 tubes, one with $q > q_c$ and one with $q < q_c$, where q_c is the value of flux which
 416 minimizes pressure drop. By computing the single eigenvalue in the discrete spectrum of
 417 the operator on the RHS of (3.17), Sampson and Gibson showed that only the former is
 418 linearly stable. *Holmes* [2007] analyzed this case in more detail by considering the full
 419 spectrum of the operator, obtaining the same results for the discrete spectrum and further
 420 showing that the continuous spectrum is exactly $\text{Range}\{E_\alpha\} = (-\infty, c)$ where $c < 0$, so
 421 that only the discrete spectrum determines the stability properties.

422

423 **Case (iii): variable pressure and flux.** This case is considerably more difficult to
 424 analyze analytically, and we will ultimately rely on numerical results. These show that as
 425 in the constant pressure case, the continuous spectrum appears to be $\text{Range}\{E_\alpha\}$ which
 426 is entirely negative, so we focus our analysis on the discrete spectrum.

427 Returning to (3.17), the equations can be rewritten by noting that α_0 solves the
 428 equation $E(\alpha_0(q, \chi)) = I(q, \chi)$, so taking the partial q -derivative and evaluating at

429 q_0 gives $\left. \frac{\partial I}{\partial q} \right|_{q_0} = \left. \frac{dE}{d\alpha} \right|_{\alpha_0} \left. \frac{\partial \alpha_0}{\partial q} \right|_{q_0}$. Here we introduce symbols $A_q, P_q, P_\alpha[\alpha_1]$ to represent the

430 derivative terms, which are defined precisely in Table 2. Under these transformations,
 431 (3.17a) and (3.17b) become

432

433
$$\frac{d\alpha_1}{dt} = E_\alpha \left(\alpha_1 - \frac{p_1 - P_\alpha[\alpha_1]}{P_q} A_q \right)$$

434

435
$$\frac{dp_1}{dt} = \frac{-\tau}{P_q} (p_1 - P_\alpha[\alpha_1])$$

436

437 To find the eigenvalues in the discrete spectrum, we look for a solution of the

438 form $(\alpha_1, p_1) = e^{\lambda t} (\tilde{\alpha}_1, \tilde{p}_1)$, substitute into the above equations, and solve to get

439
$$\tilde{p}_1 = \frac{\tau P_\alpha[\tilde{\alpha}_1]}{(P_q \lambda + \tau)} \tag{3.18}$$

440

441
$$\tilde{\alpha}_1 = \frac{-E_\alpha(\tilde{p}_1 - P_\alpha[\tilde{\alpha}_1]) A_q}{P_q(\lambda - E_\alpha)} = \frac{P_\alpha[\tilde{\alpha}_1] E_\alpha A_q \lambda}{(P_q \lambda + \tau)(\lambda - E_\alpha)} \tag{3.19}$$

442

443 These equations are valid provided $\lambda \neq -\tau/P_q$ and $\lambda \neq E_\alpha(\chi) \forall \chi$. The first is a

444 single point, which can be ignored. The second exception requires $\lambda > \max_\chi (E_\alpha(\chi))$,

445 which is simply the condition that λ is greater than the supremum of the continuous

446 spectrum, which again we denote by c . Therefore, we consider (3.18) and (3.19) only

447 for $\lambda \in (c, \infty) \setminus \{-\tau/P_q\}$.

448 Applying the operator P_α to (3.19) leads to an equation for λ :

449

450
$$F(\lambda) \equiv P_\alpha \left[\frac{E_\alpha A_q \lambda}{(P_q \lambda + \tau)(\lambda - E_\alpha)} \right] - 1 = 0. \tag{3.20}$$

451

452 If $\tau = 0$ this equation is exactly the constant-pressure case mentioned above. For
453 other values of τ we solved this equation numerically for λ in (q_i, τ) parameter space.
454 The full regions of stability/instability and oscillating solutions for a representative value
455 of dimensionless temperature constant $T_n = 10$ are summarized in Figure 6b. Let us
456 describe these in more detail.

457 Consider a fixed q_0 such that it is less than the flux q_c that minimizes pressure
458 drop. If $\tau = 0$ there is one eigenvalue, and the tube is unstable. As τ increases, there is
459 a critical value of τ at which a bifurcation occurs and the system has 3 eigenvalues. One
460 of these is real and the other two are complex with non-zero imaginary parts. The real
461 root is always negative and less than $-\tau / P_q$, so we track the signs of the complex roots
462 in order to detect instability. As τ increases, the real parts of the roots decrease,
463 eventually crossing zero so the system becomes stable. As τ is further increased, the
464 complex eigenvalues eventually disappear.

465 For $q_0 > q_c$ the system is always stable. As τ increases, a similar bifurcation
466 occurs, with complex eigenvalues appearing for large τ and disappearing for even larger
467 τ . For a fixed value of τ , this means that there is a critical value of q_0 below which the
468 system is unstable, and above which the system is stable. This critical value is plotted
469 with diamonds in Figure 6a for several values of τ . The figure shows the critical value
470 decreases as τ increases, so that the range of stable steady states is much greater with
471 large τ . As $\tau \rightarrow \infty$, all steady states become stable, corresponding to case (i) with
472 constant flux. As anticipated, the value of τ serves the function of interpolating between
473 constant pressure drop and constant flux for quantifying a stability criterion.

474

475 **3.4. Numerical Simulations of stability**

476

477 Numerical simulations of the nondimensional equations (3.15) were performed to
478 test the linear stability predictions. The pressure difference was either kept constant, or
479 varied according to (3.15b), and the tube radius was stepped forward in time using
480 (3.15a). Time derivatives were calculated using forward Euler, the trapezoidal rule was
481 used for integration, and 1000 eigenvalues were used to calculate the heat flux and steady
482 profiles. 40 points were used to represent the tube in the horizontal. The simulations were
483 stopped if the tube froze shut, i.e. when $\alpha(\chi, t) = 0$ for some χ . The numerical
484 simulations confirm the theoretical predictions. Small perturbations to a profile that is
485 linearly stable return to the original state, whereas perturbations to a profile which is
486 linearly unstable eventually freeze shut for $\tau \neq 0$. The perturbation oscillates about the
487 steady-state as it grows or decays exactly where linear theory predicts complex
488 eigenvalues.

489 Consider now the fixed-pressure case, $\tau = 0$, which is unique as it has two
490 possible steady-states, one stable and the other unstable. Figure 7 shows the two different
491 types of evolution that are possible if we start with the linearly unstable profile and
492 perturb it a little. If the perturbation is mostly positive, in the direction of the stable
493 profile corresponding to the same value of Δp , then the tube opens up, and moves to the
494 stable profile. If the perturbation is mostly negative, away from the stable profile, then
495 the tube freezes shut. As the tube moves from one profile to another, its shape is always

496 close to that of a steady profile. Any localized disturbances to the profile are rapidly
497 ironed out. This is consistent with the linear theory, which predicts large negative
498 eigenvalues in the continuous spectrum that appear to be associated with highly localized
499 eigenfunctions.

500 Figure 8 shows two cases of the radius at the endpoint of the tube $\alpha(1)$ in the
501 case of a growing or decaying oscillating solutions. The time constant τ was kept
502 constant, and the flux varied so that it was to the right of the critical flux in one case, and
503 to the left in the other. In the first case, a small perturbation oscillated about the steady-
504 state and eventually decayed, leaving a steady-state tube in its wake. In the second case, a
505 small perturbation oscillated about the steady-state but grew larger, and eventually the
506 tube froze shut.

507

508 **3.5. Application: length of a lava tube**

509
510 One motivation for this study was to explain the length of lava tubes observed in
511 some volcanic flows on Earth and Mars, where tubes of 50-200km have been found
512 [Sakimoto and Zuber 1998]. Such steady-state tubes, which are formed when highly
513 viscous lava flows down low-angle slopes, often terminate because of geographical
514 features such as an abrupt change in slope or reaching an ocean, and it would be
515 interesting to know whether there are physical constraints governing their lengths as well.
516 Therefore, as a final note, we would like to show some simple calculations to illustrate
517 how this model can be used to provide an upper bound for the length of a melt conduit in
518 an Earth or planetary context. In many tubes, the pressure at the upstream end of the tube

519 is dominated by the hydrostatic pressure so we use this as the constraint. Recalling that
 520 the non-dimensional pressure drop must be greater than a critical value in order for a
 521 steady-state tube to exist, the length satisfies

$$522 \quad \frac{\Delta Pr_0^4}{4\kappa\mu L^2} \geq \Delta p_c(T_n) \quad \Leftrightarrow \quad L \leq \sqrt{\frac{\Delta Pr_0^4}{4\kappa\mu\Delta p_c(T_n)}}. \quad (4.1)$$

523 Using typical lava parameters [*Keszthelyi 1993, Sakimoto and Zuber 1998*]

524 $\kappa = 10^{-7} \text{ m}^2 / \text{s}$, $\rho = 2300 \text{ kg/m}^3$, $\mu = 60 \text{ (54-160) Pa}\cdot\text{s}$, $T_i = 1133\text{-}1187 \text{ }^\circ\text{C}$,

525 $T_s = 1077^\circ\text{C}$, $T_0 = 30^\circ\text{C}$, (these temperatures correspond to $T_n = 8 - 20$), and

526 calculating the hydrostatic pressure difference as $\Delta P = \rho g H$, where H is the total

527 vertical distance travelled by the lava tube and g is gravity, we find that a tube with a

528 radius of 10m which drops 1km can have a maximum length of 110 - 440 km.

529 It is encouraging that this is consistent with observations, but we note that there are many

530 reasons (not detailed here) why this model is too idealized to make direct conclusions

531 about lava tubes. We note also that our mixed upstream condition applies in certain

532 flows, such as when the lava tube drains from a lava lake or an interior elastic magma

533 chamber, each receiving lava either steadily or impulsively from a source inside the earth,

534 in which case the linearized version of the geophysical upstream condition is similar to

535 3.15b. However, due to the difficulty of obtaining accurate data for such flows we prefer

536 not to speculate on numerical values at present.

537

538

539 **4. Summary and Discussion**

540

541 We conducted a laboratory experiment which shows that when fluid flows
542 through a tube whose boundary is held below freezing, solid material forms on the
543 boundary, leaving an inner tube of flowing liquid. As the flow rate is progressively
544 decreased, the pressure drop across the tube first decreases and then increases before
545 finally the tube freezes shut.

546 We investigate a theoretical model for low-Reynolds number flow through a tube
547 with solidification, in which we solve for the shape of a steady-state tube as a function of
548 distance downstream and find the relationship between pressure drop and flux in steady-
549 state. This shows that the pressure drop has a minimum as flux is varied. The linear
550 stability of the steady-states depend on the upstream boundary condition: when constant
551 flux is applied, all states are predicted to be stable; when constant pressure drop is
552 applied, those corresponding to a flux less than the flux at the minimum are unstable, and
553 for a coupled condition the critical flux for stability is in between. In the experiments,
554 pressure drop and flux were coupled by the measuring device, so these qualitative results
555 may explain the experimental rise in pressure drop as flux is slowly decreased to freezing
556 value.

557 Attempts to produce a full quantitative comparison between the laboratory
558 experiment and the theory have produced poor results which we attribute to numerous
559 possible causes of uncertainty in the experiment. There was, of course, uncertainty in the
560 mean values as well as internal variations of viscosity and specific heat, which makes
561 quantitative comparison difficult. There is also an overall sensitivity of the system to the
562 precise tube geometry, which is not captured by an axisymmetric model. Our experiments
563 showed that small perturbations near the endpoint could initiate large-scale freezing

564 events. Most solidifying materials have some crystal structure that might generate local
565 flaws, and even small bits of foreign material (particles, dust, microbubbles, etc.) might
566 produce effects that get magnified near the exit. It is possible that experiments using pure
567 filtered or distilled water that is completely free of dust, particles and dissolved air, could
568 produce results much closer to theory since it has very well-known material properties
569 and minor viscosity changes near freezing. However, it is important to note again that the
570 earlier experiments with water [e.g. *Zerkle and Sunderland*, 1968 *Mulligan and Jones*,
571 1976 *Hirata and Ishihara* 1985, *Weigand et al.*, 1997] exhibited wave formation in the
572 ice and that such local features might be common and that their role in freeze-up is
573 probably not yet fully appreciated. To clarify such points, optical views of the liquid tube
574 interiors would be very useful.

575 Overall, our findings suggest that the distance traveled by fluid in a melt conduit
576 is very sensitive to the conditions that govern pressure and flow rate at the upstream end.
577 One of our motivations was to study the paths of magma and lava flows, which are well
578 known to be quite complicated. We suggest that the sensitive interrelation between
579 upstream pressure and the stability of the tube at the downstream end, where it is most
580 likely to freeze shut, is one mechanism responsible for such complexity.

581

582 **Acknowledgements**

583 Support was received from the Geophysical Fluid Dynamics Program, which is
584 supported by the Ocean Sciences Division of the National Science Foundation under
585 Grant OCE-0325296, and from the Oceanography Section of the Office of Naval
586 Research under Grant N00014-07-1-0776. The laboratory experiments were supported by

587 the Deep Ocean Exploration Institute of W.H.O.I. M.C. Holmes-Cerfon would like to
588 thank Lou Howard for many helpful conversations during the GFD summer program. We
589 are also very grateful for the thorough help and comments of two anonymous referees.

590

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644 **Tables**

A	Upstream reservoir cross sectional area
A_n	Coefficients of temperature solution
E	Radial heat flux in the solid
G_n	Coefficients of solution for flux of temperature $A_n \phi_n'(1)$
H	Elevation of liquid upstream of the tube
I	Radial heat flux in the liquid
L	Tube length
L_H	Latent heat of solidification
P	Pressure
ΔP	Pressure drop across entire tube
Pr	Prandtl number
Q	Volume flux through the tube
Q_i	Volume flux into upstream reservoir
S	Stefan number $L_H/C_p(T_i - T_s)$
T	Temperature in the liquid
T_e	Temperature in the solid
T_i	Temperature of fluid at inlet
T_n	Dimensionless temperature constant, equal to $(T_s - T_0)/(T_i - T_s)$
T_0	Temperature at the outer radius
T_s	Temperature of solidification
a	Radius of the solid-liquid interface
c_p	Specific heat
g	Acceleration of gravity
k	Thermal conductivity
p	Dimensionless pressure
Δp	Dimensionless pressure drop $r_0^4 \Delta P / 4 \mu \kappa L^2$
$\Delta p_c(T_n)$	Critical value of pressure drop, below which no steady-state tube is possible
q	Dimensionless flux $2Q / \kappa \pi L$
q_i	Dimensionless inlet flux $2Q_i / \kappa \pi L$
$q_c(T_n)$	Critical value of flux, at which $\Delta p = \Delta p_c$
r	Radial coordinate
r_0	Outer tube radius
t	Time
u	Fluid velocity
u'	Dimensionless fluid velocity
v	Velocity in a radial direction
x	Coordinate along the axis of the tube
α	Dimensionless radius of solid-liquid interface a/r_0

ε	Amplitude of perturbation
η	Dimensionless radial coordinate r/a
θ	Dimensionless liquid temperature $(T - T_0)/(T_i - T_s)$
θ_e	Dimensionless temperature of solid $(T_e - T_0)/(T_i - T_s)$
κ	Thermal diffusivity $k/\rho c_p$
λ_n	Eigenvalues of Graetz problem
μ	Dynamic viscosity
ν	Kinematic viscosity μ/ρ
ρ	Density
τ	Pressure time constant $\pi S r_0^6 \rho g / 8 A \mu \kappa L$
ϕ_n	Eigenvectors of Graetz problem
χ	Dimensionless coordinate along axis x/L

645

646 Table 1 List of symbols. For perturbation theory, the basic state is denoted by subscript 0
 647 and the perturbation by subscript 1.

648

$$E(\alpha(\chi, t)) = \frac{\partial \theta_c}{\partial \eta} = \frac{T_n}{\ln \alpha}$$

$$I(\chi, q) = \frac{\partial \theta}{\partial \eta} = \sum G_n e^{-\lambda_n^2 \chi / q}$$

$$E_\alpha(\chi) \equiv \frac{1}{\alpha_0} \frac{dE}{d\alpha} \Big|_{\alpha_0} = \frac{-T_n}{\alpha_0^2 (\ln \alpha_0)^2}$$

649

$$P_q \equiv \frac{\partial \Delta p}{\partial q} \Big|_{\alpha_0, q_0} = \int_0^1 \frac{1}{\alpha_0^4} d\chi$$

$$P_\alpha[\alpha_1] \equiv \frac{\delta \Delta p}{\delta \alpha} \Big|_{\alpha_0, q_0} [\alpha_1] = q_i \int_0^1 \frac{-4\alpha_1}{\alpha_0^5} d\chi$$

$$A_q \equiv \frac{\partial \alpha_0}{\partial q} \Big|_{q_0} = \alpha_0 \frac{T_n}{\left(\sum G_n e^{-\lambda_n^2 \chi / q_i} \right)^2} \sum G_n \frac{\lambda_n^2 \chi}{q_i^2} e^{-\lambda_n^2 \chi / q_i}$$

650

Table 2. Particular form of the functions used in the perturbation calculations.