STEPS TOWARDS RATIONAL MODELLING OF CERAMIC INJECTION MOLDING

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An attempt is made to set up the fundamentals for applying mathematical process modelling on the basis of rational thermomechanics to the forming process of oxide ceramic pastes plasticized by thermoplastic binders. Using a continuum description throughout we start with the local balance equations known from traditional continuum mechanics and mention the special status of the entropy inequality. Looking for a material model as simple as possible but as complex as needed, constitutive theory is used to deduce a simple fluid model (the generalized Newtonian liquid) with the help of constitutive principles, and for the one-dimensional case the Herschel-Bulkley model is suggested. In order to make the theory liable to experimental verification, idealizations are made concerning flow geometry. Boundary conditions are shortly discussed. Apart from delivering a rational framework for the modelling of ceramic injection molding, the methodological approach applied in this paper should be of value also for other widely used forming processes in ceramic technology, especially cold extrusion, extrusion at elevated temperature or hot extrusion.

INTRODUCTION

Injection molding is one of the most versatile techniques for the automized large-scale production of small-sized complex-shaped ceramic parts. The whole production process from the raw materials to the final part consists in mixing and compounding the ceramic powder with the organic binder mix (binder, plasticizer and other processing aids), the forming process proper (injection molding sensu strictu), ejection of the green part, debinding (burnout of binder, plasticizer and all other organic compounds) and firing, where the ceramic body undergoes sintering in the same way as bodies formed by any other ceramic shaping technique.

The subject of the present paper is the forming process itself, where in practice a thermoplastic ceramic paste is injected into a colder metal mold cavity, in which it solidifies (under a certain hold pressure) to a rigid ceramic green body. Apart from inhomogeneities remaining after mixing and compounding, and apart from defects arising during the subsequent step of debinding, the flow pattern during injection itself is decisive for the formation of microstructure. And in contrast to the other phenomena which can be considered as faults - or the result of having not yet managed the respective process step - the formation of orientation textures in the case of non-spherical particles, gradients of density (particle packing) or other smooth changes in microstructure are essential parts of the process step itself and as such unavoidable. It seems to us that controlling this microstructure with its unavoidable "inhomogeneities" should be one of the main objectives on the way to make process control effective and to enable the production of injection-molded ceramics with reproducible properties.

While most of the work done in the field of ceramic injection molding is based on empirical approaches (cf. e.g. [10, 13, 12, 17, 14]) we feel that it is necessary to encourage research on a rational basis and to investigate the possibilities of such approaches for our purpose. Without doubt the mathematical modelling of a real ceramic injection process (i.e. in the way in which it is performed in practice) is a highly non-trivial enterprise, not to say hopelessly difficult.

The difficulties consist first of all in the material concerned. An injection-molding paste is a mixture of at least one solid and one liquid component (constituent) or phase. The solid phase for itself would behave as a flowing powder system, the organic liquid phase usually as a visco-elastic liquid. From a philosophical point of view it would therefore be most satisfactory to model such a system by multiphase mixture theory [25] and to account for the possibly visco-elasto-plastic behavior of such a material. It is clear however that in engineering practice it can never be the aim of a material model to describe all possible features of material behavior in any thinkable situation. Much more elegant is a simple model that incorporates exactly those features which are of interest for the intended applications. Thus the choice of an appropriate material model is always a compromise between realistic generality, practical usefulness and mathematical simplicity and has to be guided by a portion of pragmatism.

Not less severe is the complexity of the process itself (it is neither steady nor isothermal) and the possible flow geometries which could be of interest for ceramic injection molding [17]. As far as these points are concerned it is reasonable to make certain useful idealizations which make the process model a little less general but give way to considerable simplifications of the governing field equations and allow analytical solutions, explicit calculations and the comparison with experimental results.

SINGLE BODY CONTINUUM DESCRIPTION OF CERAMIC PASTES

A typical ceramic injection-molding mix consists of a continuous fluid phase (usually a thermoplastic binder) and a dispersed solid phase (the ceramic powder)[1]. Being interested in the gross behavior of such a material (e.g. a thermoplastic paste with an oxide ceramic powder as a filler) during the forming process, especially the velocity profile which develops during flow, we choose a continuum description for the material as a whole and do not differentiate between the individual phases (constituents). Thus we consider the material as a single body[2] characterized by a non-zero density[3], which is a smooth[4] function of the spatial position **x** and the time instant t:

$$\rho \equiv \rho(\mathbf{x}, t) > 0 \quad . \tag{1}$$

In order to describe mechanical phenomena, we introduce a further smooth field called motion by the deformation function χ, which is a function of the referential position **X** and the time instant t

$$\mathbf{x} = \chi(\mathbf{X}, t) \tag{2}$$

and define the deformation gradient

$$\mathbf{F} \equiv \mathrm{Grad}\chi(\mathbf{X}, t) \tag{3}$$

(where "Grad" denotes the referential gradient), the velocity

$$\mathbf{v} \equiv \dot{\chi} = \frac{\partial \chi(\mathbf{X}, t)}{\partial t} \quad , \tag{4}$$

the velocity gradient grad **v** (where "grad" denotes the spatial gradient) and its symmetric part called rate of deformation tensor:

$$\mathbf{D} \equiv \tfrac{1}{2}\,(\mathrm{grad}\mathbf{v} + (\mathrm{grad}\mathbf{v})^T) \quad , \tag{5}$$

where the superscript T denotes the transpose of a tensor. To account for thermal effects we would have to introduce also the temperature field

$$T = T(\mathbf{x}, t) \tag{6}$$

and its gradient.

BALANCE EQUATIONS AND ENTROPY INEQUALITY

Accepting the single body approach we have to consider the following set of balance equations known from classical continuum mechanics (here given in local form):

Mass balance:

$$\dot{\rho} + \rho\, \mathrm{div}\mathbf{v} = 0 \tag{7}$$

Linear momentum balance:

$$\rho\dot{\mathbf{v}} = \mathrm{div}\mathbf{T} + \rho\mathbf{b} \tag{8}$$

Internal energy balance:

$$\rho\dot{u} = \mathrm{tr}(\mathbf{TD}) + \mathrm{div}\mathbf{q} + Q \quad , \tag{9}$$

where ρ is the density, **v** the velocity, **T** Cauchy's stress tensor, **b** the external body force, u the internal energy, **D** the rate of deformation tensor, **q** the heat flux vector and Q the heat source. In these equations "tr" denotes the trace of a tensor, "div" the (spatial) divergence and a superimposed dot the material time derivative of a scalar or vectorial quantity φ defined by:

$$\dot{\varphi} = \frac{\partial \varphi}{\partial t} + \mathbf{v} \,.\, \mathrm{grad}\varphi \tag{10}$$

In the above case of a non-polar material (this was assumed a priori) the angular momentum balance reduces to the statement that the stress tensor is symmetric and need not be considered explicitly. The body force **b** and the heat source Q being adjustable from outside and thus considered to be known a priori, this is a system of 5 equations in 15 unknowns (ρ, τ, u, three components of **v** and **q** respectively, six components of **T**), so that it is generally necessary to close the system by specifying 10 of these dependent variables (usually the six stress tensor components, the three heat flux vector components and internal energy) by constitutive equations. Examples of very simple constitutive equations would be Newton's relation for linearly viscous fluids, Fourier's relation for heat conduction and the energetic equation of state for an ideal gas.

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[1] In contrast to some other authors even in the field of rational mechanics we prefer to call systems of this type, where the solid loading is near to the critical powder volume concentration (CPVC), "pastes " and use this handy word as a short hand synonym for the rather clumpsy expression "highly concentrated suspension". It should be clear however that principally rational approaches are apt to fit mixtures of arbitrary concentrations.

[2] In contrast to mixtures which are treated as "multiple bodies" in rational thermomechanics, cf. [25].

[3] Viz. the bulk density of the paste as a whole.

[4] I.e. as often continuously differentiable as needed.

Entropy and the Second Law have a special status in modern continuum thermodynamics (rational thermodynamics) which has been extensively dealt with in works of the Truesdell-Coleman-Noll school and many other works originating from or influenced by it, cf. e.g. [5, 15, 21, 25, 26]. In contrast to traditional thermodynamics one generally adopts the idea of considering the Second Law as a constitutive principle (called entropy or dissipation principle) which imposes restrictions on the constitutive equations and thus limits not the kind of processes which are allowed to occur but the kind of response by which a material reacts on the influences it undergoes during such a process.

The most universally accepted local form of the Second Law is the so-called Clausius-Duhem inequality in the form proposed by Truesdell and Toupin [25, 27]:

$$\rho\dot{s} + \mathrm{div}\frac{\mathbf{q}}{T} - \frac{Q}{T} \geq 0 \qquad (11)$$

where s is the entropy. For reasons of convenience and practical handiness it is usual to combine it with the First Law (in its local form, i.e. the internal energy balance (9)) and to define the Helmholtz free energy

$$f \equiv u - Ts \qquad (12)$$

to obtain the so-called reduced inequality

$$\mathrm{tr}(\mathbf{TD}) - \rho\dot{f} - \rho s\dot{T} - \frac{1}{T}\,\mathbf{q}\cdot\mathbf{g} \geq 0 \ , \qquad (13)$$

where it is evident that the last three l.h.s. terms are related to heat conduction, temperature changes and changes in free energy, i.e. thermal effects in a broader sense, while the first term accounts for dissipation due to viscous effects (internal friction), i.e. mechanical work. In a purely mechanical theory or in isothermal situations one consequence of this equation is that the viscosity coefficient must always be positive for linearly viscous fluids. For further consequences of this inequality in non-isothermal situations the reader should consult [21].

THE 3-D MATERIAL MODEL

The way how a material behaves in a certain process can be described by constitutive equations, which express the dependence of quantities characterizing the material response on the fields of density, motion and temperature. The thermomechanical response of a material at a certain spatial position in a body and at a certain time would be completely characterized by the constitutive equations for the internal energy u, the entropy s, the heat flux vector $\mathbf{q}$ and the symmetric stress tensor $\mathbf{T}$. These quantities depend in general on the set of independent variables ρ, χ and T and their histories (i.e. all their former values) in the whole body. This is in rough words the meaning of a slightly extended version of the principle of determinism as known from constitutive theory. The fact that all quantities (u, s, $\mathbf{q}$ and $\mathbf{T}$) are assumed to depend on the same set of independent variables and histories and no preferences are made a priori is consistent with Truesdell's principle of equipresence (equipresence rule) [25].

From a mathematical point of view, one could write down the abstract form of such a dependence by a response functional, whose independent variables would be functions of time (from the infinite past up to the present instant t) and all material points $\mathbf{Y}$ of a body including the one under actual consideration $\mathbf{X}$. However a theory of such generality has never been developed. Almost all materials of engineering interest can be modelled as local materials, i.e. materials obeying the principle of local action. In essence it says that the near neighborhood of a material point has a stronger influence on the material point under consideration than the more distant neighborhood. Mathematically this corresponds to substituting spatial functions as independent arguments of the response functional by its local values, gradients and higher gradients at this specific material point. The corresponding principle of differential memory states something similar in dimensions of time: The near past has a stronger influence on the material response of a material point than the distant past. Mathematically this means to substitute history functions as independent arguments in the response functional by their actual values, time derivatives and higher time derivatives at the actual time instant.

With accepting these two principles we can replace the complicated response functional by a response function $\mathcal{F}$ and write the general thermomechanical material response in the following form:

$$\begin{aligned}
&\{u, s, \mathbf{q}, \mathbf{T}\} = \\
&= \mathcal{F}[\mathbf{x}, t, \rho, \chi, T, \\
&\qquad \mathrm{grad}\rho,\ \mathrm{grad}(\mathrm{grad}\rho),\ \ldots\ \dot{\rho},\ \ddot{\rho}\ \ldots\ \dot{\overline{\mathrm{grad}\rho}},\ \ldots \\
&\qquad \mathrm{Grad}\chi,\ \mathrm{Grad}(\mathrm{Grad}\chi),\ \ldots\ \dot{\chi},\ \ddot{\chi},\ \ldots\ \dot{\overline{\mathrm{Grad}\chi}},\ \ldots \\
&\qquad \mathrm{grad}T,\ \mathrm{grad}(\mathrm{grad}T),\ \ldots\ \dot{T},\ \ddot{T},\ \ldots\ \dot{\overline{\mathrm{Grad}T}},\ \ldots\ ] \ , \qquad (14)
\end{aligned}$$

where all arguments except for $\mathbf{x}$ and t itself are fields, i.e. functions of $\mathbf{x}$ (or $\mathbf{X}$) and t.

Although this equation is very general, by adopting it we have already excluded models for materials exhibiting non-local effects and for materials with a long-range memory. The first step can easily be justified, because non-local effects certainly are not relevant for the materials and processes that are interesting in ceramic technology. Slightly more intricate is the justification for ignoring long-range memory effects in ceramic injection molding, since it is well known that e.g. theories of fading memory, integral-type or rate-type materials are widely used in polymer science to describe certain phenomena exhibited by thermoplastic materials. These materials serve as typical binders for the ceramic injection molding process and it is not a priori clear, why a neglection of memory effects should be justifiable here. In fact typical thermoplastic ceramic pastes do show effects characteristic for materials with memory (e.g. the die-swell or Barus effect at tube exits). It can however not be the aim of a material model to describe a material as such in all its aspects, i.e. to predicate its possible behavior in all thinkable situations or processes. A material model should be first of all as simple as possible and therefore has to focus on those specific and typical features of behavior which are absolutely necessary for the processes to which the model is intended to apply. And from this point of view an inspection shows that long-range memory effects are of minor importance for the materials considered here and the application in mind. As far as mechanical behavior is concerned we have to describe mainly two features of ceramic injection molding pastes:

- The non-linear flow behavior (shear-rate dependent apparent viscosity) and
- the phenomenon of a yield value of stress.

Neither of these is necessarily related to long-range memory effects and later in this paper it will be evident that for reasons of experimental practice it is not useful to choose a more complicated model.

With regard to the intentions of this paper we confine the following deductions to the purely mechanical material response, i.e. to the stress tensor in the case of isothermal processes. Thus we ignore here the constitutive equation for the heat flux vector (in numerical simulations of non-isothermal flow or plastic injection molding Fourier's law is usually applied), for entropy and for internal energy, and by omitting the temperature gradient in the set of independent variables we treat the material as if it were a non-conductor of heat. Furthermore we model our ceramic injection molding paste as a so-called simple material ([18], cf.also [26] and other textbooks), i.e. we omit density gradients and higher deformation gradients in the set of independent variables. While of certain importance for mixture models [15, 21], no necessity has arisen to apply a theory of non-simple materials within the context of a single body approach and Noll's theory of simple materials [18] comprises all material models that are known from traditional and linear irreversible thermodynamics. A differential memory with respect to density has never been observed [15] and experience has shown that differential memory with respect to temperature can be ignored in processes where the temperature changes are not too abrupt (for isothermal processes it is not relevant at all), so that we can also omit the time derivatives of density and temperature in the set of independent variables. For the sake of simplicity we tentatively omit higher gradients and time derivatives. The constitutive equation for the stress tensor is then:

$$\mathbf{T} = \bar{\mathbf{T}}(\mathbf{x}, t, \rho, T, \chi, \mathrm{Grad}\chi, \dot{\chi}, \dot{\overline{\mathrm{Grad}\chi}}) \ . \tag{15}$$

It can easily be shown that a dependence of this type is the simplest possible equation which allows the description of viscous effects (internal friction), which is necessary to model real fluids in processes where they do not behave as Eulerian (i.e inviscid) ones. A more familiar form of this equation is [21] :

$$\mathbf{T} = \bar{\mathbf{T}}(\mathbf{x}, t, \rho, T, \chi, \mathbf{F}, \mathbf{v}, \mathrm{grad}\mathbf{v}) \ . \tag{16}$$

Even now the dependence is too general from a physical point of view. Some of the independent variables are excluded by the principle of material objectivity, which ensures the invariance of material response with respect to Galilei transformations of the observer. In regard of this principle the motion (deformation function) χ the velocity $\mathbf{v}$ and the skew-symmetric part of the velocity gradient as well as the explicit dependence on the spatial position $\mathbf{x}$ and the time instant t vanish. Furthermore the intended application to an isotropic fluid allows us to exploit the symmetry principle, with the help of which it can be shown that the dependence of the deformation gradient $\mathbf{F}$ is redundant [15, 21]. The remaining constitutive equation is then:

$$\mathbf{T} = \hat{\mathbf{T}}(\rho, T, \mathbf{D}) \ . \tag{17}$$

This is not all. The principle of material objectivity bears another important consequence insofar as it states than any objective tensor function (and $\mathbf{T}$ is a priori introduced as an objective quantity, cf. [21], must transform according to then transformation law of a geometrical tensor, i.e. must have following property:

$$\mathbf{QTQ}^{\mathrm{T}} = \mathbf{Q}\hat{\mathbf{T}}(\rho, T, \mathbf{D})\mathbf{Q}^{\mathrm{T}} = \hat{\mathbf{T}}(\rho, T, \mathbf{QDQ}^{\mathrm{T}}) \ . \tag{18}$$

Here $\mathbf{Q}$ denotes an arbitrary orthogonal tensor (i.e. det $\mathbf{Q} = +1$ for proper and det $\mathbf{Q} = -1$ for improper rotations). Tensor functions with this property are called isotropic [15, 21], and such isotropic tensor-valued functions with a symmetric second-order tensor argument ($\mathbf{D}$) can be written in a more explicit form without loss of generality (by the Cayley-Hamilton theorem and the representation theorem known from tensor analysis, cf. [2, 15, 26]):

$$\mathbf{T} = \phi_0 \mathbf{I} + \phi_1 \mathbf{D} + \phi_2 \mathbf{D}^2 \ . \qquad (19)$$

In this equation **I** denotes the unit tensor and the scalar coefficients ϕ_i are generally functions of ρ, T and the three principal invariants of **D** defined as:

$$I_D \equiv \mathrm{tr}\mathbf{D}$$

$$II_D \equiv \tfrac{1}{2}[(\mathrm{tr}\mathbf{D})^2 - \mathrm{tr}\mathbf{D}^2] \qquad (20)$$

$$III_D \equiv \det\mathbf{D} \ . \qquad (21)$$

For our purposes a further and very significant simplification of this equation follows from the fact that for processes of engineering interest a ceramic injection molding paste can be considered as incompressible as long as vacuum voids, air inclusions or gas bubbles are absent[5]. In this case the continuity equation for mass (7) reduces to the statement:

$$\mathrm{div}\mathbf{v} = 0 \ . \qquad (22)$$

It can be shown easily that this implies a zero value of the first invariant:

$$\mathrm{div}\mathbf{v} = \mathrm{tr}\mathbf{D} = I_D = 0 \qquad (23)$$

and a corresponding simplification of the second invariant II_D.

Furthermore the pressure p in the well-known stress tensor decomposition for simple fluids:

$$\mathbf{T} = -p\mathbf{I} + \tau \qquad (24)$$

(with τ denoting the viscous or dissipative stress tensor) does not correspond to the thermodynamic pressure any more (i.e. ceases to be determined by an equation of state) but becomes a hydrostatic pressure of arbitrary magnitude, so that the first r.h.s. term of (19) containing the unit tensor can be absorbed in it.

Thus the viscous stress tensor can be written in the following way:

$$\tau = \phi_1(\rho, T, \mathrm{tr}\mathbf{D}^2, III_D)\mathbf{D} + \phi_2(\rho, T, \mathrm{tr}\mathbf{D}^2, III_D)\mathbf{D}^2 \quad (25)$$

(where the first r.h.s. term accounts for shear stresses and the second for normal stresses as can easily be shown for simple shear flow, cf. [1]). As is well known from experimental rheology normal stress effects like the Barus effect (die-swell) or the Weissenberg effect usually appear at free boundaries. Injection molding on the other hand is essentially a process where - apart from the moving flow front in the mold cavity - free boundaries are of no importance. This fact and the regard to the intended applications of the model in the context of microstructure formation, which is realized only during the flow in fixed boundaries, give us the right to ignore the second r.h.s. term of (25). Concerning the third invariant on the set of arguments it can be shown [22] that for simple shear flows the determinant of **D** vanishes[6] and the resulting constitutive equation for the shear stress is the well known model for so-called generalized Newtonian liquids:

$$\tau = 2\eta(\rho, T, 2\mathrm{tr}\mathbf{D}^2)\mathbf{D} \ , \qquad (26)$$

the numerical factors being conventional [1]. The so-called apparent viscosity η is a function of bulk density ρ[7], temperature T and the argument that can in simple shear flows be interpreted as a square of a scalar shear rate or, equivalently, as the shear of one component of the velocity gradient.

SPECIAL FLOW GEOMETRIES AND BOUNDARY CONDITIONS

Explicit calculations of velocity profiles in general 3-D flow processes usually require a very careful analysis of the respective geometry and the choice of an appropriate numerical algorithm to solve the governing PDEs. This is a highly non-trivial task for whole branch of engineering called Computational Fluid Dynamics (CFD) and goes beyond the objective of the present paper. It is clear that for explicit calculations and predictions of velocity profiles a constitutive equation of the form (26) is still too general. Therefore one is forced to assume a certain form of the dependence of η on the argument $2\mathrm{tr}\mathbf{D}^2$, which should be plausible, simple and permit a physical interpretation of the new coefficients or parameters introduced. The general procedure to manage this problem in commercial software packages is to take one of the well known rheological models developed for 1-D situations (in the simplest case Newton's constitutive

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[5] The question whether or not a ceramic mix can be considered as incompressible is not trivial. Pure liquids and solids are commonly considered as incompressible in contrast to gases or gas-containing liquids. This is reasonable. Note however that a liquid-solid mixture can be compressible even if both phases are incompressible. This is due to possible changes in the solid volume fraction during flow. If the solid particles are not by chance neutrally buoyant, i.e. of equal density as the surrounding liquid, the mixture as a whole can well undergo changes in (bulk) density. However for the solid loadings near to the CPVC as is the case in most injection molding pastes, the assumption of incompressibility should not be too far from reality.

[6] Many different types of flow are locally simple shear flows and belong to this class, but it is out of question that in the case of mold filling in ceramic injection molding a certain care must be taken when the mold cavity is of complicated form, e.g. containing sudden contractions or expansions.

[7] Remembering the incompressibility condition, i.e. the fact that ρ is constant in the flow considered, this dependence just means the trivial statement that η depends on the type of the paste, e.g. its solid volume fraction.

equation for linearly viscous fluids) and to assume its validity for general 3-D flow.

We with follow a similar scheme here with the only difference that with respect to the intended experimental verification of computed results we apply these rheological models to the special case of unidirectional, locally simple shear flow, i.e. for the flow situation for which they were actually designed.

Let us consider unidirectional one-dimensional flow under a pressure gradient (Poiseuille flow) in a cylindrical tube with constant circular cross-section (cf. figure 1). This geometry was chosen to allow easy comparison with the capillary viscometer geometry in subsequent work [19], but the treatment of plane Poiseuille flow (i.e. unidirectional two-dimensional flow) which is also of importance in injection molding, would be entirely analogous.

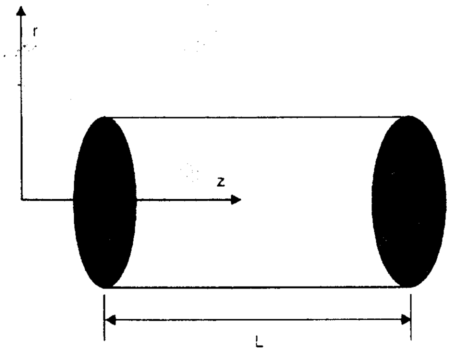

Figure 1. Flow geometry (cylindrical Poiseuille flow).

In unidirectional one-dimensional cylindrical Poiseuille flow to the radial and angular velocity components vanish, and for steady flows the axial velocity at each point is only a function of the radial position. For this situation we can define a so-called shear rate γ, which is the radial gradient of the axial velocity component:

$$\gamma \equiv \gamma(r) = \frac{\mathrm{d}v_z(r)}{\mathrm{d}r} \,. \tag{27}$$

It is easy to show that in this case (and analogically in other cases of simple shear flow) the third argument in (26) reduces to the square of the shear rate defined above:

$$2\mathrm{tr}\mathbf{D}^2 = \gamma^2 \,, \tag{28}$$

so that the apparent viscosity η in these simple shear flows depends only on density, temperature and shear rate. Thus the (one-dimensional) constitutive equation for the shear stress component τ_{rz} can be written in the form:

$$\tau \equiv \tau_{rz} = \eta(\rho, T, \gamma^2)\gamma \,. \tag{29}$$

Such a shear-rate dependence of the apparent viscosity of course is one of the most important features that can experimentally be observed for various non-Newtonian fluids and must be taken into account to describe their behavior. However, nothing has been said so far about the explicit form of this dependence. At this stage rational deduction ends. The material model is exactly determined and for the simplified situation of simple shear it is clear how experimental measurements have to proceed in principle: One has to measure the shear rate at certain point and time and the corresponding value of the shear stress in the material at this point and time. Thus from a theoretical viewpoint all further models are redundant, because it is principally possible to determine the flow curve[8] experimentally to any desired degree of accuracy [4].

For at least three reasons however it is useful to adopt one of the well-known empirical rheological models to fit the measured flow curves :

- Adopting a physically reasonable model makes interpolations (and to a certain degree extrapolations) possible and thus reduces the number of experiments needed to determine the complete flow curve in the range of interest.
- If a calculation of velocity profiles is intended, this task is substantially facilitated, when the course of the flow curve can be expressed by a simple model. In some cases analytical solutions can be obtained.
- Using an appropriate empirical model for a class of similar materials offers the possibility to collect and compare rheological data of different systems, e.g. ceramic injection molding pastes with different powder contents, with different binder systems or at different temperatures.

A very simple and widely used model in different fields of non-Newtonian fluid dynamics and rheology is the so called "power law":

$$\tau = K\gamma^n \,, \tag{30}$$

where K is called coefficient of consistency and n flow index.

For its simplicity this equation is one of the favorite equations used in commercial software packages for non-Newtonian fluids without memory and has also been applied to thermoplastic injection molding mixes [7, 9, 16].

As is well known from traditional ceramic technology a non-zero value of yield stress is characteristic for

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[8] We call a flow curve the graphical representation of the one-dimensional constitutive equation an measured by standard methods of experimental rheology, e.g. the capillary viscometer method.

classical ceramic mixes containing clay minerals and experience shows that yield phenomena occur also in ceramic injection molding mixes, The simplest way to include yield stress in the rheological model is given by Bingham's equation

$$\tau = \tau_0 + K\gamma \ , \qquad (31)$$

where τ_0 is the yield stress and K is called plastic viscosity in this context.

To describe both the non-linear flow curve and the phenomena of yield stress we prefer the three parameter Herschel-Bulkley model, which can be looked upon as a combination of the two preceding ones:

$$\tau = \tau_0 + K\gamma^n \ . \qquad (32)$$

Another model which is sometimes recommended [9] for ceramic injection molding mixes is Casson's model, which has the advantage of containing only two parameters:

$$\sqrt{\tau} = \sqrt{\tau_0} + \mu\sqrt{\gamma} \ . \qquad (33)$$

For a more complete list of the possible models the reader should consult standard textbooks on non-Newtonian rheology (e.g. [1, 22, 23, 28]). It should be noted that all coefficients (parameters) occurring in these equations are generally dependent on the density (viz, the bulk density, determined e.g. by the solid volume fraction) and on the temperature of the mix.

By inserting the constitutive equation into the linear momentum balance (or one of the one-dimensional models into the corresponding one-dimensional balance) a field equation is obtained that can be solved for certain boundary conditions in steady processes[9]. Without going into details here we think that a few remarks concerning the latter might be useful.

For cylindrical Poiseuille flow one boundary condition is trivially satisfied on grounds of the flow symmetry. It asserts that at the tube axis the velocity profile has a tangent plane which is perpendicular to the flow direction. In other words, the stress distribution is continuous at this position and the velocity profile continuously differentiable ("smooth").

The second boundary condition which is usually assumed asserts that the material does not slip at the wall during flow. In contrast to the first boundary condition mentioned above this second one is by no means obvious and would principally require experimental verification. It is generally believed, that for "normal" (i.e. pure small molecules) fluids real slip does not exist and that the occurrence of wall-slip has nothing to do with the degree of adhesion of a certain material to a specific surface[10]. On the other hand it is well known that multiphase fluids (e.g. suspensions) and polymer solutions can exhibit a phenomenon reminiscent of wall slip (the so-called "apparent" or "effective" slip) which is a consequence of local concentration variations during flow along solid boundaries [23, 3]. Moreover it seems that in polymer melts where large macromolecules are present, and the ratio of molecule size to surface roughness scale is large, real wall slip does occur [6, 24]. Principally these effects cannot be excluded in the systems which are of interest here. Experience with ceramic mixes has shown however that for specially prepared internal surfaces of capillaries (increased roughness by material abrasion) the influence of effective slip flow is so small in the systems considered that it is not measurable by standard experiments of paste rheology [11, 19]. It seems therefore reasonable to assume the no-slip condition as long as no plausible and verifiable alternative can be suggested a priori. If the mathematical flow model based on this assumption works and is in agreement with experimentally observed facts to the desired degree of accuracy, this will be a hint a posteriori that the assumption was realistic.

SUMMARY AND OUTLOOK

In this paper we presented the theoretical fundamentals for rational modelling of injection molding of thermoplastic ceramic pastes. It has been shown that the central problem at this stage is the choice of a sufficiently general and complex but reasonably simple and explicit material model (constitutive equation). This problem has been solved here by applying the principles of constitutive theory well-known from rational thermomechanics (principles of determinism, local action, differential memory, material objectivity, material symmetry, cf. e.g. [21, 25, 26]) to the case of typical ceramic injection molding mixtures and discussing their relevance for these materials with respect to the intended applications. For the sake of simplicity the treatment in the present paper has been confined to isothermal situations. For non-isothermal situations application of the entropy principle mentioned in section 3 results in certain restrictions concerning free energy and the coefficient of heat conduction if Fourier's law is adopted, but the coupled problem of solving non-isothermal flow of non-Newtonian fluids is a highly non-trivial one and remains to be a subject of future theoretical research. Certainly it is not a field for merely experimental work to be done by ceramists.

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[9] For non-steady problems initial conditions have to be considered, too.

[10] Coleman, Markovitz & Noll [4] report that even mercury does not slip along the smooth walls of a glass capillary during flow.

A further point might be worth mentioning: Throughout the paper ceramic injection molding mixes are treated by a single body continuum approach. With regard to the multiphase composition of such mixes it could be desirable to use a multiple body continuum description that takes into account the individual behavior of the respective phases. Rational mixture theories offer a reasonable way to do this [25, 20]. They are widely applied in soil science, hydrology and geology, and on their basis it is possible to get a qualitative understanding of many phenomena observed in the flow of suspensions. So far their application to paste flow is not usual and quantitative predictions based on these theories are complicated by serious difficulties in the determination of individual boundary conditions for the different phases [20].

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KROKY SMĚŘUJÍCÍ K RACIONÁLNÍMU MODELOVÁNÍ INJEKČNÍHO VSTŘIKOVÁNÍ KERAMIKY

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V tomto článku, který má tvořit teoretický základ dalších prací a navazujících článků zabývajících se experimentální stránkou problému, se pokoušíme o aplikaci racionálního přístupu na aktuální problematiku poměrně velkého praktického významu, injekčního vstřikování keramických past. Jako typický materiál si lze představit oxidový prášek s termoplastickým pojivem, i když tento článek podává poněkud širší rozhled a materiálové modely zde prezentované nevylučují jejich aplikace na jiné soustavy.

Způsob popisu se opírá o mechaniku kontinua a o racionální termomechaniku (racionální termodynamiku). Vycházíme zde z popisu kinematiky, formulujeme lokální bilanční rovnice a zmíníme se krátce o speciálním postavení entropické nerovnosti v této teorii.

Centrálním problémem racionálního modelování injekčního vstřikování je výběr vhodného materiálového modelu. Zvolený materiálový model je obecná Newtonská kapalina. Jeho výhoda spočívá v tom, že je velice jednoduchý a zároveň dovoluje popsat právě ty jevy, které nás v procesu injekčního vstřikování zajímají nejvíce: mez toku a nelineární průběh tokové křivky. Odvození tohoto materiálového modelu je prováděno krok za krokem pomocí principů konstitutivní teorie, aby bylo vidět rozsah a význam učiněných předpokladů pro případ injekčního vstřikování keramických past a je formulováno se snahou docílit maximální přesnosti vyjádření bez použití příliš složitého matematického aparátu.

Možnost experimentálního ověření modelu v následujících pracích vyžaduje určité idealizace např. ohledně geometrie toku. Pro Poiseuilleův válcový tok i jiné prosté smykové toky preferujeme z důvodů uvedených v práci Herschel-Bulkleyův model, který představuje speciální jednorozměrný případ modelu obecných Newtonských kapalin, ačkoliv se z něho nedá dedukovat pomocí racionálních principů konstitutivní teorie. Krátká zmínka je věnována složitému problému okrajových podmínek.

Kromě adaptovaného racionálního rámce pro injekční vstřikování je zde uplatněn metodologický přístup aplikovatelný i na jiné tvarovací procesy keramiky, např. extruzi.