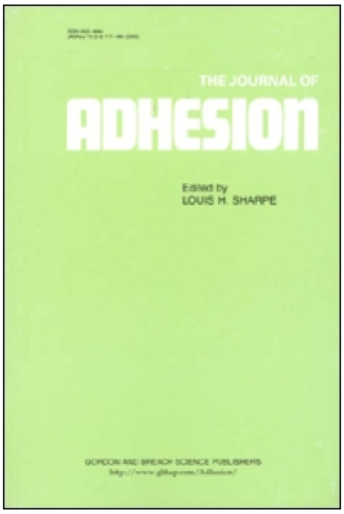

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Adhesive Dispensing Process Analysis: Steady-State Dispensing of One-Component Adhesives

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The process of dispensing one-component heat-cure adhesives was investigated in order to understand current application processes and to guide new process development. Typical one-component adhesives exhibit non-Newtonian rheological behavior, and hence Newtonian fluid mechanics does not adequately describe the dispensing process. In the present study, the adhesives were modeled as Bingham fluids possessing a yield stress and a steady state viscosity. The model of the dispensing apparatus includes four major flow sections connected in a serial configuration. The fluid mechanics equations derived for Bingham fluids in the individual flow sections were solved by numerical methods in order to understand the interrelationships between the material variables (*e.g.* yield stress, viscosity, temperature dependencies) and process variables (*e.g.* pressure, flow geometry, temperature, output). The concept of the model is generic and the details of the model can be modified for any forced-flow adhesive application process.

The adhesive flow properties significantly influence the process output. Dispensing temperature, among the process variables, has the strongest effect on process output. A ± 1.0°C perturbation in the dispensing temperature can cause as much as a 14% variation in the bead size for the range of adhesives studied. Differences in flow characteristics result in differences in processability and non-linear temperature/pressure sensitivity. The non-linear sensitivity can be eliminated by operating the dispensing process isothermally. Finally, the process limits for one-component adhesives, which are susceptible to chemical instability induced by viscous heating during processing, are defined and discussed in terms of a modified Brinkman number that takes into account viscous dissipation, heat conduction and convection, and chemical stability of the material during processing.

KEY WORDS adhesive dispensing; epoxy adhesives; non-Newtonian flow; rheology; thermal stability; viscous heating

INTRODUCTION

One-component adhesives based on filled epoxy formulations have been used or proposed for use in automotive body construction processes for structural and semi-structural bonding. Commercial one-component epoxy adhesives contain not only liquid epoxy resins with solid curatives but also fillers, plasticizers, rubber modifiers, pigments, and other additives. Their rheological behavior is rather complicated as shown in a recently completed study describing the flow properties of several commercially available adhesives.[1] The adhesives were found to exhibit three major classes of flow characteristics: low yield stress and viscosity, high yield stress and low viscosity, and high yield stress combined with high viscosity. When adopting these and similar materials for automotive manufacturing, it is essential that their flow properties be compatible with application equipment and process conditions.

A typical adhesive dispensing system consists of an adhesive tank, a pump, connecting pipes, a material conditioner, a flow control valve, and an extrusion nozzle (Figure 1). The geometry of each of the components in the flow path influences the volumetric flow rate of the adhesives. The process output (volumetric flow rate of the adhesive) is also sensitive to changes in the process variables such as

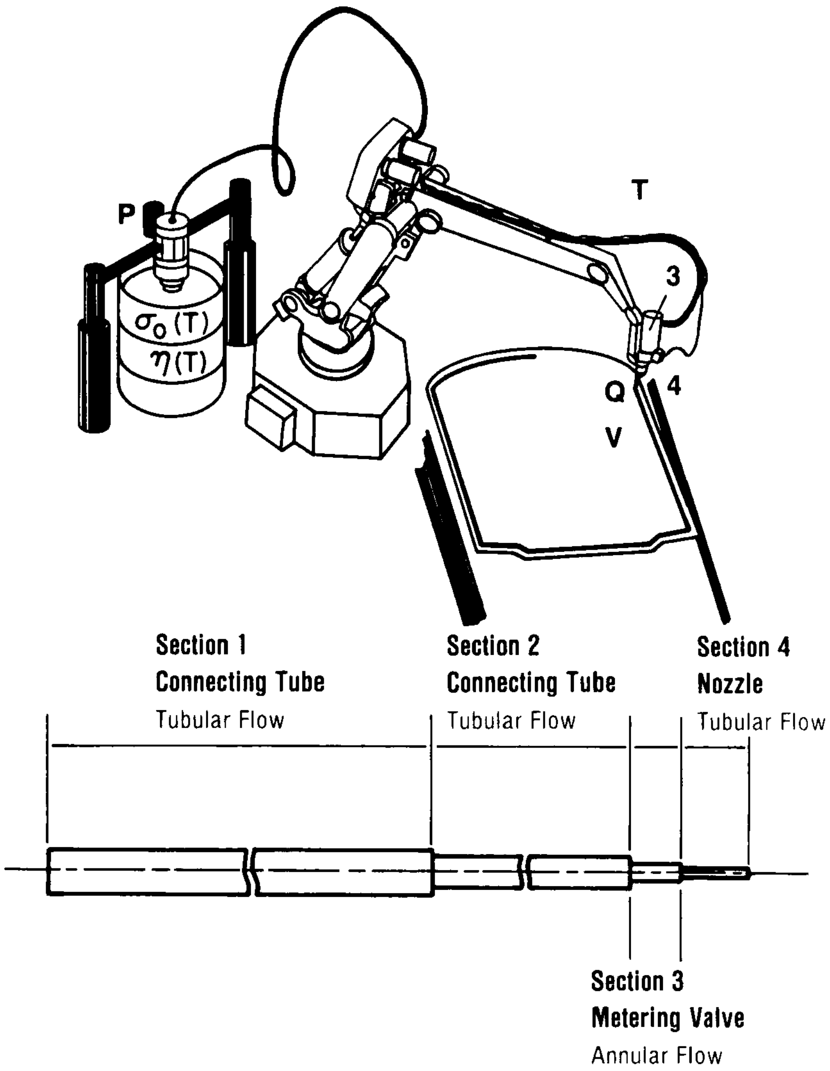

FIGURE 1 Schematic of a typical adhesive dispensing process for automotive manufacturing process.

temperature and pressure. The interrelationships between the material flow properties, the dispensing equipment, and the process variables must be investigated to understand the dynamics of the adhesive application process. Another important aspect of adhesive dispensing is chemical stability. The viscosity of one-component adhesive pastes can increase dramatically during processing due to chemical crosslinking reactions if excessive viscous heating is not avoided.

Conceptually, the flow path of the adhesive paste can be sectioned and simplified such that a set of fluid-mechanics equations can be written to represent mathematically the viscous flow of the adhesive through the dispensing hardware.[2,3] Thus, the relationships between the dispensing system variables (*e.g.* viscosity, yield stress, flow path geometry, pressure and temperature) can be studied by analyzing the solutions of such a set of fluid-mechanic equations. This report describes the results obtained from a systematic investigation of the adhesive process by numerically solving the fluid-mechanics equations with actual material properties and process variables obtained from a typical production process. The quantitative results obtained are thus specific to the process modeled. Qualitatively, however, the results are generic, and the discussion focuses on trends between the material and process variables applicable to any forced-flow dispensing process of adhesive pastes. The process sensitivity is defined and discussed in detail. Viscous-heating induced chemical instability of the one-component adhesives is analyzed by using an energy equation. A set of process limits is also defined in terms of material and process variables.

METHODS AND RHEOLOGICAL DATA

1 Fluid Mechanics

A schematic of a typical adhesive dispensing process for automotive closure panel production is shown in Figure 1. The adhesive flow in the dispensing apparatus was divided into four major sections: 1 and 2 are tubular flow connecting tubes (9.525 mm (⅜ in) and 3.175 mm (¼ in) ID), 3 is an annular flow metering valve, and 4 is a tubular flow final nozzle. If an adhesive bead of radius R_b and length l_b is to be produced by the dispensing process within a cycle time t_c, the amount of adhesive material flowing through the system, defined as the volumetric flow rate of the process (process output), is:

$$q = \frac{\pi R_b^2 l_b}{t_c} \tag{1}$$

where q designates the process flow rate. A set of process parameters was recorded from a typical hem-flange adhesive dispensing process. The cycle time, bond-line length, and bead diameter were 3.8 sec, 2261 mm (89 in), and 3.175 mm (⅛ in), respectively. The desired volumetric flow rate was found to be 18.5 ± 0.2 ml/sec. These process parameters were used in the study to illustrate the model calculations.

Since all the adhesive pastes possess an apparent yield stress, Newtonian fluid mechanics is inadequate to describe the adhesive flow profile in each section of the process hardware. The simplest flow model that incorporates the yield behavior is the Bingham model of fluid flow in which the flow is described by two parameters: a yield stress, σ_o, and a Newtonian viscosity, η; $\sigma = \sigma_o + \eta\gamma$, where σ and γ are shear stress and shear rate, respectively. Therefore, the Bingham model is used in this study to represent the adhesive paste properties. The tubular flow of a Bingham material is described analytically by the Buckingham-Reiner equation:[4]

$$q_i = \frac{\pi \Delta P_i}{8\eta l_i} R_i^4 \left[1 - \frac{4}{3}\left(\frac{\sigma_o}{\tau_1}\right) + \frac{1}{3}\left(\frac{\sigma_o}{\tau_1}\right)^4 \right] \tag{2}$$

$$\tau_1 = \frac{\Delta P_i}{l_i} R_i \tag{3}$$

where q_i, R_i, l_i, and ΔP_i designate the volumetric flow rate, radius, length, and pressure drop in a tubular section i, respectively. The flow of adhesives through the metering valve (for example, a needle valve) can be approximated by annular flow. Bird and Frederickson derived an analytical expression relating volumetric flow rate to pressure drop of a Bingham fluid in an annulus:[5,6]

$$q_v = \frac{\pi \Delta P}{8\eta l_v} R_v^4 \left[(1-\kappa)^4 - 2\lambda^2(1-\kappa)^2 - \frac{4}{3}(1+\kappa^3)\beta + \frac{1}{3}(4\lambda^2 + \beta^2)^{3/2}\beta \right] \tag{4}$$

where q_v, ΔP, R_v and l_v designate flow rate, pressure drop, radius and length of the needle valve, respectively. The dimensionless rheological parameter β, the annulus geometry parameter κ, and the boundary layer thickness ratio λ are defined as:

$$\beta = \frac{2\sigma_o l_v}{\Delta P R_v} \tag{5}$$

$$\kappa = \frac{R_{needle}}{R_v} \tag{6}$$

$$2\lambda(\lambda - \beta)\ln\frac{\lambda - \beta}{\lambda\kappa} - 1 + (\beta + \kappa)^2 + 2\beta(1 - \lambda) = 0 \tag{7}$$

Equations 4 to 7 can be used to describe the flow of an adhesive paste (Bingham fluid) through a needle valve typically used for controlling the material flow rate. The size of the gap between the needle and the valve seat, which is related to κ, is calculated from the outer and inner radius of the annulus R_v and R_{needle}.

The flow of adhesive paste through the dispensing apparatus is represented by connecting individual components in a serial configuration. The flow rate in each section is equal to the total flow rate (similar to electric current),

$$q = q_1 = q_2 = \cdots\cdots = q_i \tag{8}$$

and the sum of the pressure drops of each section (similar to electric potential) is equal to the total pressure drop,

$$\Delta P_{total} = \sum \Delta P_i \tag{9}$$

TABLE I
Summary of flow path geometry and processing parameters for process modeling

Connecting Pipe 1:	length = 7315 mm (24.0 ft), diameter = 9.525 mm (⅜ in)
Connecting Pipe 2:	length = 1219 mm (4.0 ft), diameter = 6.35 mm (¼ in)
Flow Control Valve:	length = 15.0 mm $R_v = 2.5$ mm, $R_{needle} = 0.2$ to 2.4 mm
Nozzle:	length = 25.4 mm (1.00 in), diameter = 3.175 mm (⅛ in)
Cycle Time:	3.8 sec
Desired Bead Size:	3.175 mm (⅛ in)
Bond Line Length:	2159 mm (85 in)
Pressure Range:	68.95 to 6895 kPa (10 to 100 psi)
Temperature Range:	15°C to 45°C

Therefore, the dynamics of the dispensing process can be modeled by matching the flow rate and calculating the pressure drops, in much the same way that an electric circuit is modeled. In this study, a FORTRAN program was written to solve the set of equations using a simple numerical approach.[7] The upper and lower limits of each process or material parameter are listed in Table I. In the numerical calculations, one variable at a time was allowed to change while others were held constant. The effects of changes in viscosity, yield stress, pressure, and temperature were studied.

2 Adhesive Flow Properties

A detailed characterization of the rheological properties of the one-component adhesives was reported in a previous study.[1] Interested readers are referred to that report for the rheological behavior of the adhesive pastes. Four important flow parameters: apparent yield stress, plastic viscosity, activation energy of yield stress, and activation energy of viscosity, reported in the previous investigation, are used in this study. These parameters of the one-component adhesives with identical sample names used in previous study are listed in Table II. The flow parameters describe the flow characteristics of the filled pastes over the temperature range typical of the automotive manufacturing environment. The parameters were originally extracted

TABLE II
Summary of flow properties of one-component epoxy adhesives[1]

Adhesive sample	Yield stress, Pa at 25°C (77°F)	Viscosity, Pa.s at 25°C (77°F)	E_η, kcal/mole	E_σ, kcal/mole
ES-1	56.2	39.5	23.72	7.614
ES-2	97.1	30.2	25.61	2.825
ES-3	313	11.6	29.30	6.919
ES-4	144	22.5	20.44	5.735
ES-5	174	155.2	24.92	7.119
ES-6	273	22.1	22.41	3.875
ES-7	413	329.5	20.06	4.170

[1] Yield stress (σ_0) and viscosity (η) determined by fitting flow curves to the Casson equation, $\sigma = (\sigma_0^{1/2} + (\eta\ \gamma)^{1/2})^2$

E_η and E_σ designate activation energies of viscosity and yield stress respectively.

from rheological measurements by using the Casson equation.[1] Although the Casson equation differs from the Bingham model in the low-shear-rate region, there is negligible difference in the adhesive behavior predicted by the two equations when the shear rate is higher than about 10 sec^{-1}. We can safely use these parameters, therefore, for our fluid-mechanics calculations since the shear rates in dispensing applications are on the order of 10 to 100 sec^{-1} or higher. Time-dependent shear thinning effects (thixotropy) did not play a large role in the flow behavior of the pastes studied, and these effects have not been included in the present work.

RESULTS AND DISCUSSION

1 The Difference Between a Newtonian and a Non-Newtonian Fluid for Dispensing Applications

Traditionally, the commercial paste adhesives for many industrial applications have been characterized with a single Newtonian viscosity index measured under a forced flow arrangement. The Newtonian viscosity index generally provides an adequate description of the flow of typical un-filled adhesives but is oversimplified and can lead to erroneous results when used for process engineering calculations for filled materials. Figure 2(a) shows the difference in flow profiles when pumping a Newtonian and a Bingham fluid through a straight pipe (the simplest flow geometry). The Newtonian fluid exhibits a parabolic flow profile while the Bingham paste

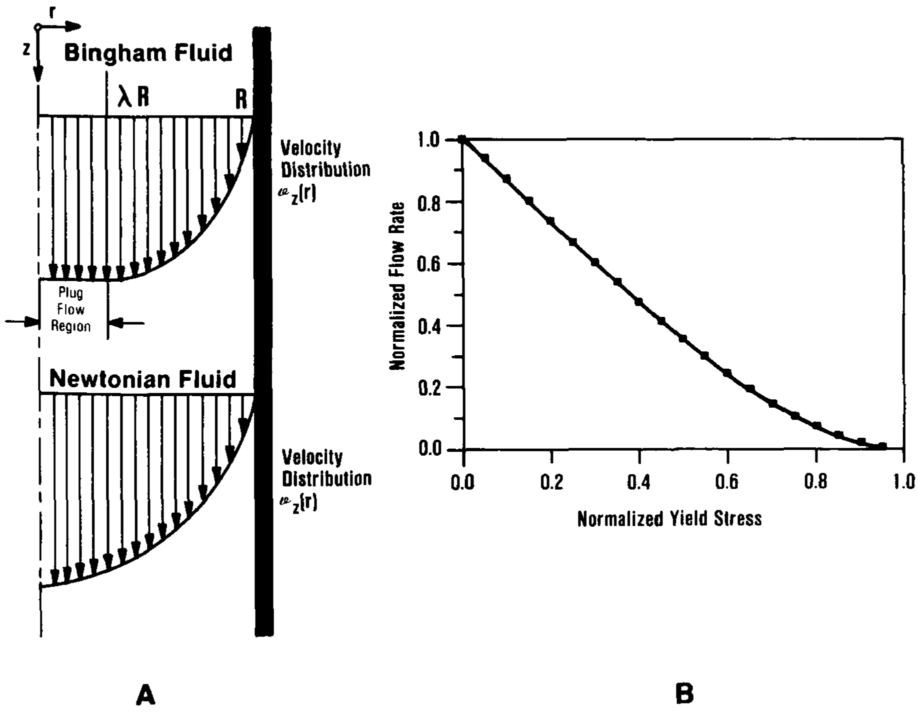

FIGURE 2 Tubular flows of a Newtonian fluid showing a parabolic velocity distribution and a Bingham fluid with a "plug" shape profile.

shows a "plug flow" velocity distribution. A lower flow rate is observed in the Bingham fluid due to the additional flow resistance attributed to the yield component. The ratio of the flow rates of a Bingham fluid over a Newtonian liquid with identical viscosity is plotted *versus* the ratio of yield stress with respect to the pressure drop normalized to the tube geometry.[2] The ratio of flow rates approaches zero as the ratio of yield stress to pressure drop approaches unity. When the pressure drop is insufficient to overcome the yield stress, the Bingham fluid stops flowing. Therefore, the effect of non-Newtonian behavior on adhesive processing becomes stronger at high yield stress or low pressure drop. The non-Newtonian contribution from the yield phenomenon on steady-state flow rate is not insignificant even at the shear rates for high-speed paste dispensing. If a simple Newtonian flow model were employed to predict the output of a dispensing process, there would be a 15 to 40% over-estimate of overall flow rate as compared with the results generated using the Bingham model.

2 Effect of Yield Stress

The effect of yield stress on the overall flow rate is examined in Figure 3, in which the flow rate is plotted against yield stress. The flow rate is shown decreasing at higher yield stress, primarily due to the changes in the velocity profile in the flow path. As yield stress increases, the velocity distribution changes from a parabolic

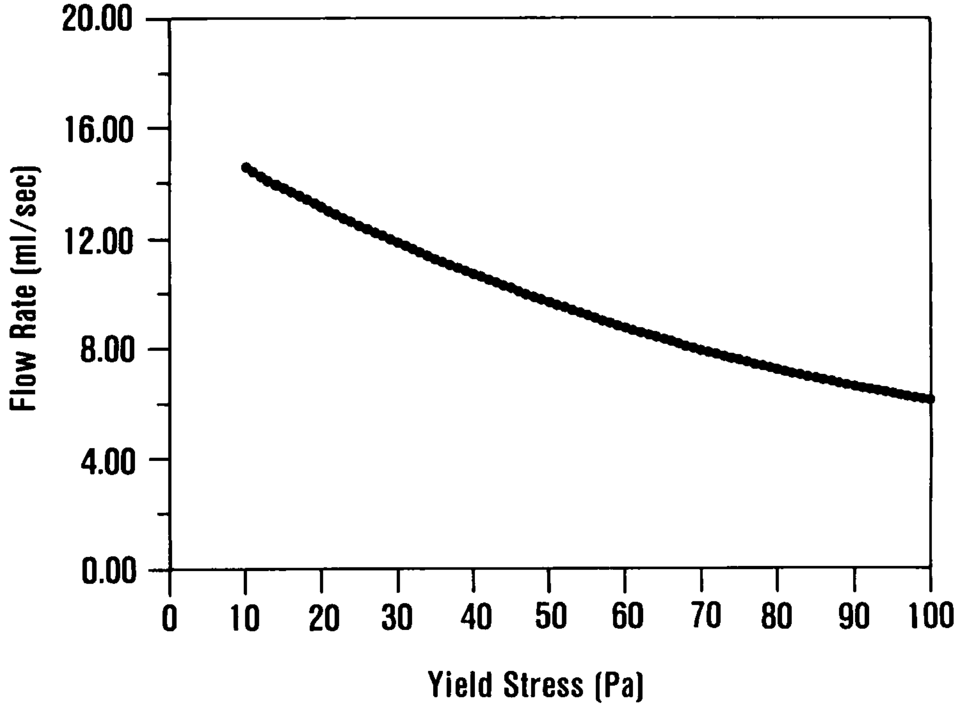

FIGURE 3 Effect of yield stress on process output measured as volumetric flow rate.

shape to a so called "plug flow" profile. The shear rate, which is zero in the flat region of the flow profile, increases very rapidly near the tube wall. Mathematically, the near-wall shear rate value,γ_w, in each section of the flow path can be obtained by taking the derivative, $(\partial V_z/\partial r)_{p,\sigma}\cdots$, of the velocity distribution function $V_z(r)$: for tubular flow:

$$\gamma_w = \frac{\partial V_z}{\partial r_{r=R}} = \frac{\Delta P}{2\eta l}R + \frac{\sigma_o}{\eta} \tag{10}$$

and for annular flow:

$$\gamma_w = \frac{\partial V_z}{\partial r_{r=R_v}} = \frac{\Delta P R_v}{2\eta l_v}[\beta + \kappa - \lambda^2 \ln\kappa] \tag{11}$$

The near-wall shear rate for tubular flow (in connecting pipes) and for annular flow (in the metering valve) can be calculated from Equations 10 and 11, respectively. The near-wall shear rate is an important parameter in determining the extent of viscous heating in a Bingham fluid since the local viscous heat generation is proportional to the square of the local shear rate which is highest near the tube wall.[2]

3 Effect of Pump Pressure

Figure 4 shows an approximately linear relationship between volumetric flow rate and pump pressure when other variables such as temperature and material properties are held constant. There is, however, a critical flow resistance in the paste that

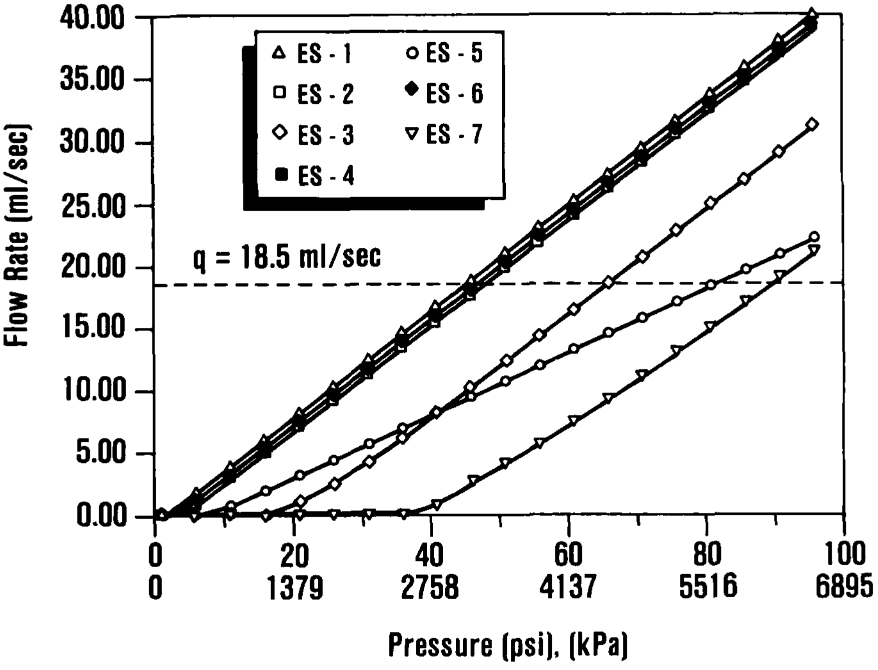

FIGURE 4 Effect of pump pressure on process output measured as volumetric flow rate.

must be overcome by a minimum pump pressure to generate a flow. The minimum pressure is proportional to the yield stress of the adhesive. The proportionality constant between the pressure and the flow rate is related to both the paste viscosity and the yield component because the flow profile changes with pressure. The adhesive pastes (ES-1, ES-2 and ES-6) with low yield stress and viscosity flow easily and possess similar pressure/flow rate relationships. It is worth mentioning that a linear relationship between pressure (control variable) and flow rate (to be controlled) simplifies process and quality control provided other variables remain constant. Ideally, all adhesive pastes to be dispensed should have similar flow characteristics allowing processing using similar control parameters. In this sense, the numerical simulation of the dispensing process offers a significant advantage in screening adhesive pastes for a particular application by using data obtained from standard rheological measurements. The high-viscosity, high-yield-stress paste ES-7 exhibited the highest resistance to flow, as indicated by a high pump pressure at a given flow rate. A different set of process parameters and a much larger metering valve are needed to dispense this type of adhesive paste using current process hardware. The ES-5 and ES-3 pastes possess either a high-viscosity or a high yield stress, but can still be processed by the typical dispensing device simulated here. The control parameters, however, must be adjusted to accommodate their unique flow characteristics.

4 Effect of Dispensing Temperature

The extremes of temperature variation in a non-air-conditioned plant environment range from about 18 °C to 40 °C. Since the viscosity and yield stress of the adhesive pastes depend upon temperature, the dispensing system must be able to compensate for changes in viscosity and yield stress of the paste and still generate a controlled flow rate within the tolerance specification. In our previous study, the temperature dependence of the flow characteristics of the adhesive pastes was represented by using the activation energy of an Arrhenius equation. To simulate the sensitivity of the volumetric flow rate of a particular adhesive paste to the temperature, the Arrhenius equation is used again to represent the change of yield stress and the viscosity due to temperature change. The variation of the paste flow rate due to the changes in viscosity and yield stress must first be studied before a proper control strategy can be organized. Figure 5 shows the relationship between the flow rate and the dispensing temperature from 15 °C to 45 °C, while pressure and flow geometry remain constant. The steady-state flow rate is very sensitive to the dispensing temperature due to the highly temperature dependent flow parameters of the adhesive pastes. As the temperature increases, both the viscosity and the yield stress of the pastes decrease and the flow rate increases. The yield stress, however, decreases at a slower rate than the viscosity due to its lower activation energy. Therefore, the yield stress has a greater contribution in governing the paste flow rates at higher temperatures. The overall effect of temperature on the flow rate, which appears to be linear on the semi-log scale, can be mathematically represented as $q = q_0 \exp \alpha T$ (q_0 and α are constants) in the temperature range examined. The constant α contains weighted contributions from the viscous and the yield components and can be calculated by linear regression analysis of the curves in Figure 5.

The process output, measured by the volumetric flow rate, is very sensitive to

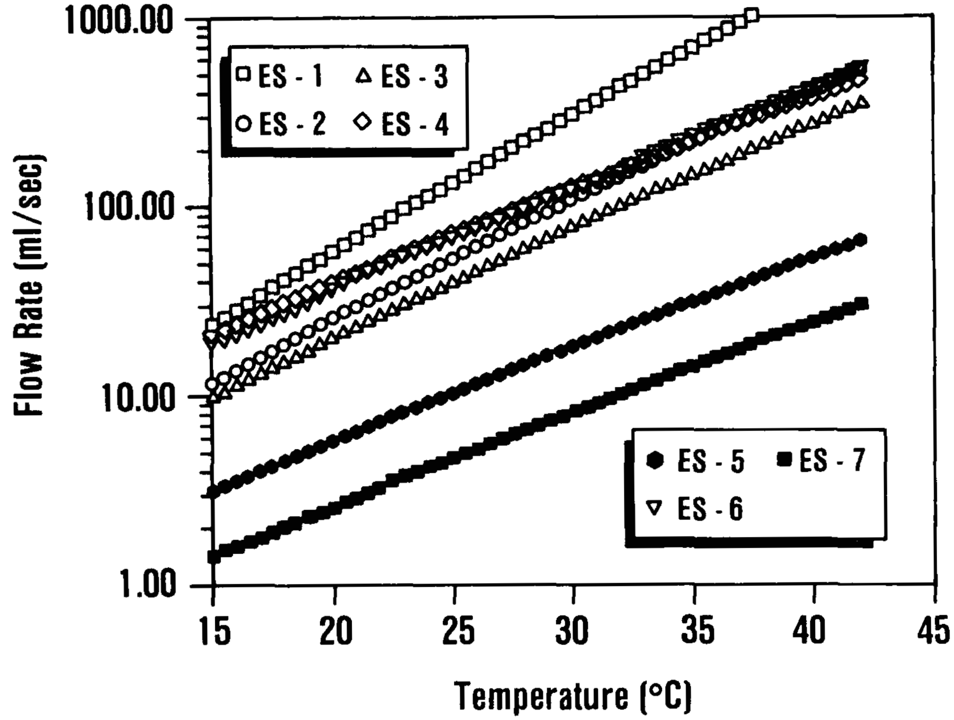

FIGURE 5 Effect of dispensing temperature on process output measured as volumetric flow rate.

the dispensing temperature as shown as Figure 5. A small perturbation in dispensing temperature can cause a rather large change in flow rate if not compensated for by adjusting other process parameters. The temperature sensitivity of the process output can be measured by the partial derivative of flow rate q with respect to temperature T (temperature sensitivity = $(\partial q/\partial T)_{p,r}$), in this case $\partial q/\partial T = q_0\alpha \exp \alpha T$. This equation is useful in estimating flow variations which would occur for a given temperature fluctuation. The equation also shows that the temperature sensitivity of flow rate itself is a function of temperature. For example, a flow rate variation of ±5.42 ml/sec/°C is calculated for the ES-1 adhesive if the temperature variation is ±1.0 °C at 25 °C dispensing temperature. This corresponds to a 0.44 mm (±14.1%) variation in the size of the adhesive bead for the ±1 °C temperature perturbation. If the temperature is controlled within ±0.1 °C, the bead size variation will only be ±0.3% for this adhesive at the 25 °C processing temperature.

5 Process Sensitivity

Once the relationships between the flow rate and each of the material and process variables become clear, we would like to know the cross-relationship between the process variables (temperature, pressure, flow geometry) while maintaining a constant flow rate. Such a cross-relationship can be calculated numerically by searching for the given flow rate while systematically varying both process variables. Figure 6 shows the relationship between the pump pressure and the dispensing temperature at a constant volumetric flow rate ($q = 18.5$ ml/sec) for four different materials, respectively. The relationships differ from approximately linear in material ES-7 to inverse proportionality in material ES-1. Furthermore, each material seems to have different offset constants due to its rheological parameters. For example, the ES-1 material offers the widest operating range (from 18 °C to 37 °C) while the ES-7 material can only be processed at higher end of the temperature and pressure range (from 34 °C and up). The temperature/pressure sensitivity of the dispensing system is indicated by the slope of the temperature/pressure curve, $\partial T/\partial T$, for a given adhesive paste. As shown in Figure 6, the pressure/temperature relationship and the system sensitivity are both temperature dependent over the operating range. The non-linear relationship between the two process variables (temperature and pressure) implies that it is difficult to operate a non-isothermal dispensing process first because the flow rate is very sensitive to temperature as shown previously, and second because it takes a different amount of pressure change to compensate for temperature fluctuations at different operating temperatures. Furthermore, all the process control parameters must be readjusted when switching from one material to another since each material responds differently to the process. When the dispensing process is operated in an isothermal condition, it is much easier to achieve a constant flow rate since the process sensitivity, $\partial P/\partial T$, can be linearized

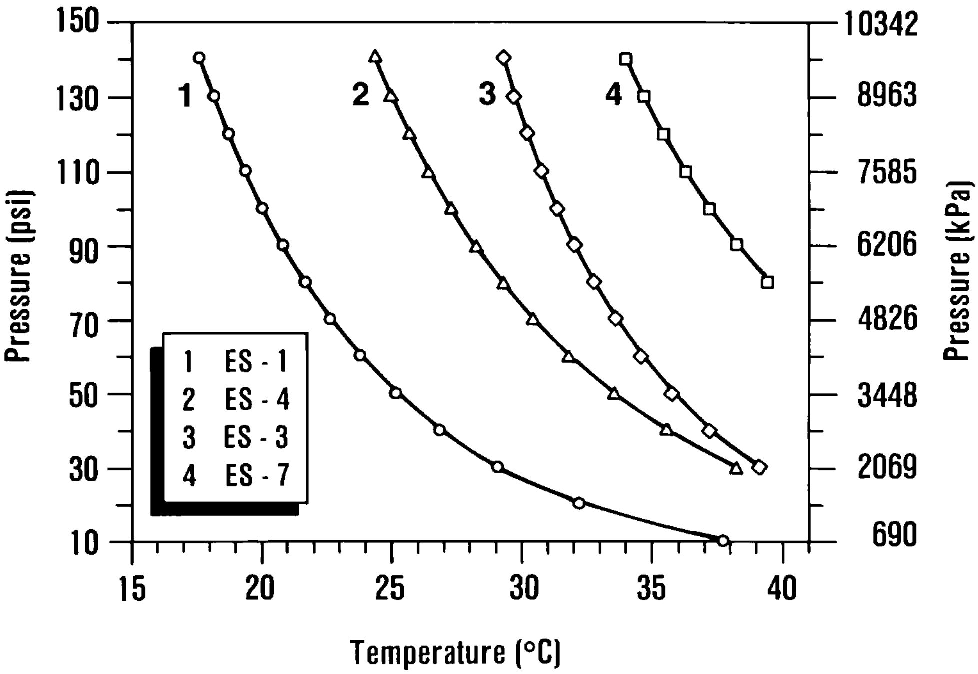

FIGURE 6 Process sensitivity between dispensing temperature and pressure at a constant volumetric flow rate (process output) $q = 18.5$ ml/sec.

around the operating temperature. A similar argument holds true for the relationship between the relative valve opening and temperature.

6 Process Limits

There are many design, equipment, and material constraints when a one-component adhesive dispensing process is considered for an automotive manufacturing application. Once the selection of material, equipment and process output is made, a set of process limits is defined. There are absolute process limits (physical limits) that cannot be exceeded simply due to the mechanical design of the equipment, for example, the maximum operating pump pressure, maximum opening of a flow control device, or the shortest cycle time of the robot. These limits are rarely approached in the actual operation. There is another type of process limit due to the chemical or physical instability of the materials. For example, solidification of the paste adhesive in the process hardware has been reported.[8] Most one-component epoxy adhesives, when subject to high shear rates, are not indefinitely stable within the operating temperature range because the viscous heating caused by high shear rates can induce cross-linking reactions in the formulated epoxy resins. The cross-linking reactions result in a dramatic increase in viscosity and ultimately transform the liquid paste into a solid Therefore, it is desirable to establish a process limit below which the flow-induced chemical instability is not an issue.

There are three major aspects of the flow-induced chemical instabilities during processing of one-component adhesives: 1, the flow-induced viscous dissipation; 2, the heat transport characteristics of the flow; and 3, the chemical reaction kinetics reflected by the sensitivity of the material to temperature. A thorough analysis of this problem requires setting up and solving the continuity, momentum, energy, and reaction kinetics equations simultaneously. Due to the complex nature of these sets of equations, such an attempt is beyond the scope of this study. We have evaluated the problem using simplified equations and dimensional analysis to gain insight into this rather complex problem.

When an adhesive paste is forced to flow in a tube, the shearing of the paste results in the generation of heat. Conceptually, the flow can be regarded as involving the slipping of a nested set of cylindrical shells in which each cylindrical shell rubs against an adjacent shell. This rubbing together of adjacent layers of fluid produces heat, which is to say that mechanical energy is steadily transformed to thermal energy. The volume heat source resulting from this viscous dissipation is usually designated by S_v. Its magnitude depends on the local velocity gradient (shear rate); the more rapidly adjacent layers move with respect to each other, the greater will be the viscous dissipation. In the tubular flow of a Bingham fluid, the shear rate is higher near the tube wall and the viscous dissipation is also more severe in this region (see Fig. 7). The rate of local viscous dissipation of a Bingham fluid flowing in a tube is given by:[3,9]

$$S_v = -\tau_{rz}(dV_z/dr) = \sigma_o\gamma_w + \eta\gamma_w^2 \tag{12}$$

where τ_{rz} and γ_w are shear stress and the near-wall shear rate, respectively. As shown in the equation, the rate of viscous heat generation, S_v, is related to both the

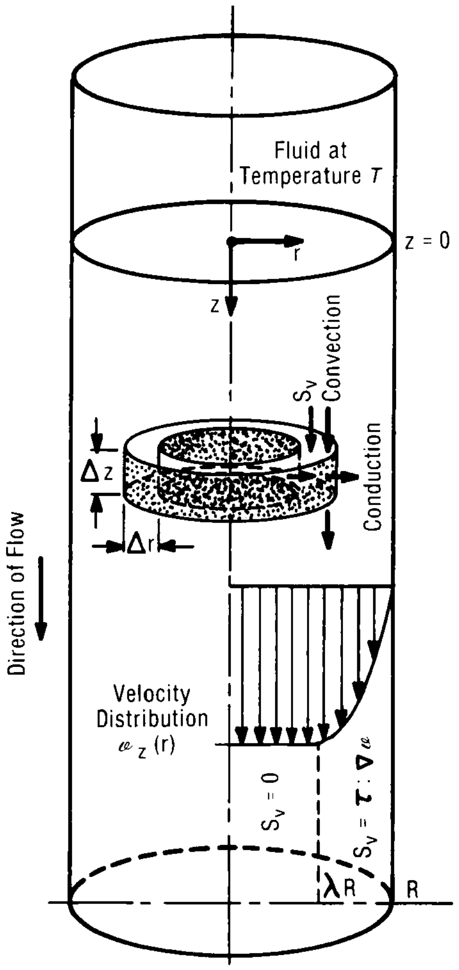

FIGURE 7 Energy balance in a Bingham fluid flowing through a tube.

material variables (yield stress and viscosity) and the process variables (shear rate at a given pressure and temperature). One can obtain an explicit relationship between S_v and other variables by substituting Equations 10 into 12 for tubular flow:

$$S_v = \frac{\sigma_o \Delta PR}{2\eta l} + \frac{\sigma_o^2}{\eta} + \eta\left(\frac{\Delta PR}{2\eta l} + \frac{\sigma_o}{\eta}\right)^2 \tag{13}$$

and by substituting Equation 11 into 12 for annular flow:

$$S_v = \sigma_o\left[\frac{\Delta PR_v}{2\eta l_v}(\beta + \kappa - \lambda\ln\kappa)\right] + \eta\left[\frac{\Delta PR_v}{2\eta l_v}(\beta + \kappa + \lambda\ln\kappa)\right]^2 \tag{14}$$

These equations indicate that the rate of viscous heat generation S_v is proportional to the square of yield stress and pressure drop, and inversely proportional to the viscosity. The true dependence, however, is much more complicated since the pressure drop itself depends upon the material and process variables, such as the viscosity and the flow rate. Equations 13 and 14 allow the rate of viscous heat generation to be quantified, provided that the pressure drop in each flow section is already calculated from numerical simulation of the dispensing process. Table III lists the calculated pressure drops at 25 °C for each of the adhesives in the particular flow geometry described in Figure 1. Table III shows that a very large pressure drop occurs in the flow control valve (section III). As a result of the large pressure drop, a very large near-wall shear rate (1458 $\sec^{-1}$) is calculated in the flow control valve for one of the materials (ES-1). The local viscous heat generation rate near the wall is 2.2 J/(ml sec) at this shear rate when Equation 14 is used. Since both the pressure drop and the near-wall shear rate are lower, the viscous heat generation rate in the connecting tube is much lower, only 0.2 J/(ml sec), under the same process conditions. Thus, viscous heating is most likely to occur in the flow control valve or near the wall of a small orifice where high shear rates are intrinsic.

The heat produced by viscous dissipation causes the temperature to rise in the paste unless it is transferred away by heat conduction. The temperature rise in the paste could cause the adhesive paste to polymerize if the onset temperature of the cross-linking reaction is exceeded. For a given selection of material, equipment, and output combination, there is a process limit below which the material is not heated significantly by viscous dissipation and is stable for practical and engineering considerations. We now proceed to define and calculate the contribution of viscous dissipation for a generic process and apply it to define the process window.

The temperature rise in the paste is determined by the balance between the viscous heat generation and the heat transport characteristics. An energy balance over the flow section is needed to quantify the temperature profile in a flow section.[3] There are three important terms in the energy equation; 1, the viscous dissipation term; 2, the conduction term; and 3, the axial temperature rise (convection) term. For a Newtonian fluid with constant heat capacity, C_p, and thermal conductivity, k, the energy equation is:

$$\rho C_p(v\cdot\nabla T) = -k\nabla^2 T + \eta(\nabla v)^2 \tag{15}$$

Axial Temperature Rise = − Conduction Loss + Viscous Dissipation

TABLE III
Pressure drops and Brinkman numbers for one-component adhesives under typical process conditions

Adhesive sample	P1, KPa (psi)	P2, KPa (psi)	Onset temp of cure, °C	ΔT, °C	Brinkman number Br′
ES-1	131 (19.0)	172 (25)	96	74	0.82
ES-2	134 (19.5)	193 (28)	124	99	0.71
ES-3	207 (30.0)	241 (35)	80	55	1.81
ES-4	131 (19.0)	179 (26)	81	56	1.22
ES-5	248 (36.0)	317 (46)	135	110	1.60
ES-6	128 (18.5)	172 (25)	78	53	1.25
ES-7	303 (44.0)	317 (46)	102	77	4.94

P1: total pressure drop in section 1 and 2 (pipes)
P2: pressure drop in section 3 (flow control valve)

A complete analysis of the viscous dissipation in the flow is achieved by solving this equation for a given flow geometry and a set of boundary conditions. In the cases where a complete analytical solution is not needed, dimensional analysis can usually provide sufficient information to sort out the important contributions to the system.[3] The advantage of using dimensional analysis is that one can find out whether viscous dissipation is important or not without solving the differential equation for the detailed temperature distribution in the flow system. Brinkman showed that by choosing a proper set of reduced variables, the energy equation is transformed to a dimensionless form with an important dimensionless parameter, termed the Brinkman number, which governs the ratio of viscous dissipation to heat conduction in a Newtonian fluid flowing in a tube:[3,9]

$$Br = \frac{\eta(V/D)^2}{k(\Delta T)/D^2} \tag{16}$$

$$Br = \frac{\mathit{viscous\ dissipation}}{\mathit{heat\ conduction}} \tag{17}$$

where D, V, k, and ΔT designate the diameter of the tube, the average flow velocity, the thermal conductivity of the fluid, and the temperature difference between the fluid and the wall. A low value of Brinkman number means that any heat produced by viscous dissipation can be readily transferred away by heat conduction. The heat generated by viscous dissipation is just balanced by the conduction when the Brinkman number is unity. A Brinkman number greater than 1 indicates there is a temperature rise in the fluid due to viscous heating. The Brinkman number, as defined in Equation 16, can only be applied, however, to the Newtonian fluids for which it was derived.[9]

In order to perform the dimensional analysis for viscous dissipation in a Bingham fluid where high shear rates occur near the wall of a tube or an annulus, we write the energy equation for the spatial element near the wall, see Figure 7:

$$\rho C_p v_z \frac{\partial T}{\partial z} = -k\frac{\partial^2 T}{\partial r^2} + \sigma_o\left(\frac{\partial V_z}{\partial r_{r=R}}\right) + \eta\left(\frac{\partial v_z}{\partial r_{r=R}}\right)^2 \tag{18}$$

The term on left side designates the temperature rise in the axial (z) direction. The first term on the right side governs the rate of heat removed by conduction in the radial direction. The rate of viscous dissipation in a Bingham fluid is represented by the last two terms. This equation applies only to the Bingham fluid in the high-shear region from λR to R where high shear rates occur. A steady state flow assumption was also made in writing the equation. Following Brinkman and Bird,[3,9] a dimensionless number, Br, for a Bingham fluid can now be obtained from the dimensional analysis performed using Equation 17 over the high-shear region of the "plug" flow in a tube:

$$Br = \frac{\dfrac{\sigma_o \Delta P R}{2\eta l} + \dfrac{\sigma_o^2}{\eta} + \eta\left(\dfrac{\Delta P R}{2\eta l} + \dfrac{\sigma_o}{\eta}\right)^2}{\dfrac{k\Delta T}{(1-\lambda)^2 R^2}} \tag{19}$$

where R(1 − λ) is a characteristic length in a 'plug flow' (Bingham fluid) where viscous dissipation is significant due to high shear rates. The velocity of the adhesive near the tube wall is also much lower than that in the "plug flow" region, therefore, the adhesive residence time near the tube wall is much longer. The temperature difference, designated as ΔT, is defined as the temperature rise in the fluid as a result of viscous heating in the high shear region. By taking the temperature rise in the axial (z) direction into consideration, the dimensional analysis can be taken one step further. The axial temperature rise is represented by the dimensionless group, $\rho C_p V \Delta T/l$, where ρ, C_p, and V are the density, heat capacity, and near-wall average velocity of the paste. As stated previously, ΔT designates the temperature difference between the dispensing temperature and the temperature of the fluid element subject to viscous heating. When the axial temperature rise (convection) term is included in the analysis, a dimensionless quantity that ratios the viscous heating to the sum of the conductive and convective heat transport is defined as:*

$$Br' = \frac{\dfrac{\sigma_o \Delta P R}{2\eta l} + \dfrac{\sigma_o^2}{\eta} + \eta\left(\dfrac{\Delta P R}{2\eta l} + \dfrac{\sigma_o}{\eta}\right)^2}{\dfrac{k\Delta T}{((1-\lambda)^2 R^2)} + \dfrac{\rho C_p V \Delta T}{l}} \tag{20}$$

$$Br' = \frac{viscous\ dissipation}{total\ heat\ transport} \tag{21}$$

The redefined Brinkman number, Br′, can be used to assess how important viscous dissipation is in an adhesive dispensing system for a given process condition represented by the process and material variables. A lower Brinkman number is the result of higher thermal conductivity, lower flow resistance and shorter paste residence time in the high-shear region.

The final step in defining the process limits for adhesive dispensing is to take into account the chemical instability of the adhesive pastes induced by the viscous dissipation. The onset of instability for the one-component adhesives corresponds to the beginning of cross-linking. For the present calculations, we have defined a reaction onset temperature, T_r. The reaction onset temperature T_r depends on the chemical composition of the material and is related to the chemical kinetics of the cross-linking reaction. The lower T_r, the more reactive (and the less stable) the adhesive. Operationally, we have used differential scanning calorimetry to determine T_r; the onset temperature has been taken to be the temperature at which the exotherm characteristic of cure is first observed.

To introduce the chemical instability issue into the Brinkman number, the temperature difference ΔT in Equation 19 is defined as the difference between the dispensing temperature T and the onset of crosslinking reaction T_r:$\Delta T = T_r - T$. Once ΔT is defined this way, the redefined Brinkman number becomes a criterion for evaluating the flow-induced chemical instability of the adhesive paste during processing. A Brinkman number lower than unity indicates the temperature rise in

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*The dimensionless ratio of viscous heating to total heat transport could also be obtained by writing a macroscopic energy balance over the layer of fluid elements near the tube wall. The result obtained from an energy balance is the same as that shown here.

the adhesive paste will not reach the onset temperature of cross-link reaction, T_r; therefore, the adhesive is processable. If the Brinkman number is equal to or greater than unity, the temperature rise induced by viscous heating in the adhesive is likely to reach or exceed the onset temperature of the cure reaction. The adhesive paste may start to solidify around the wall of the flow path, causing a further increase in viscous heating due to a narrower flow path and a higher shear rate. Once the flow-induced adhesive cure is started, the phenomenon is self-reinforcing and can cause potential processing problems. If Br' is found to be greater than 1 for a given condition, the process parameters must be revised to make it less than unity. This sets a new process limit: the Brinkman number Br' calculated from the processing and material variables must be equal to or less than one. The Brinkman number Br' for a given adhesive decreases with an increase in the diameter of the flow path (connecting tube or the metering valve) or a decrease in the length of the connecting tube, provided a constant process output (flow rate) is maintained. The shortest possible flow path from the pump to the dispensing nozzle is generally desired in order to reduce the viscous dissipation and the residence time of the adhesive.

The Brinkman number itself could be used to rank the chemical instability of any one-component thermoset adhesive subject to a forced flow dispensing process. The process limit defined here is generic and, with appropriate definition of T_r, can be applied to the dispensing of any one-component adhesive that possesses an inherent chemical instability. To illustrate the use of the redefined Brinkman number Br' in determining the material's chemical stability, we now proceed to calçulate the redefined Brinkman number for each of the adhesive pastes at a selected process condition ($q = 18.5$ ml/sec and $T = 25$ °C) to rank their susceptibility to viscous heat generation. The onset temperature of the crosslinking reactions, T_r, for each of the adhesive pastes, determined by differential scanning calorimetry,[1] is listed in Table III. The temperature difference between dispensing temperature and T_r is calculated for each material. Literature values for heat capacity (1.25 J/g °C), density (1.9 g/ml), and thermal conductivity (0.0069 J/sec cm °C) of filled epoxy resins were used for the calculation.[10] The results are also listed in Table III.

Flow-induced chemical instability of the one-component adhesive pastes during processing can be compared by ranking their Brinkman numbers. The adhesives that possess a low Brinkman number, *e.g.* ES-1 and ES-2, are chemically stable during processing due to their low resistance to flow as well as their high curing temperatures. Another group of adhesives (ES-3, ES-4, and ES-6), which exhibit a higher extent of chemical instability measured by lower T_r's, is more sensitive to viscous dissipation as indicated by their higher Brinkman numbers. The higher Brinkman numbers are the result of a small difference between dispensing temperature and onset of thermoset cure, ΔT. One of the adhesives, ES-7, which has the highest Brinkman number as a result of the highest flow resistance and an intermediate T_r, is strongly subject to viscous dissipation under the processing conditions investigated. Careful design and equipment selection is necessary to process this material in order to avoid high shear rates.

Once the modified Brinkman number, Br', calculated for a specific selection of adhesive and process, is known, the operating range of the process can be defined by adjusting the process variables (*e.g.* pressure, temperature, or size of the metering valve) such that the Brinkman number is lower than unity. In this case,

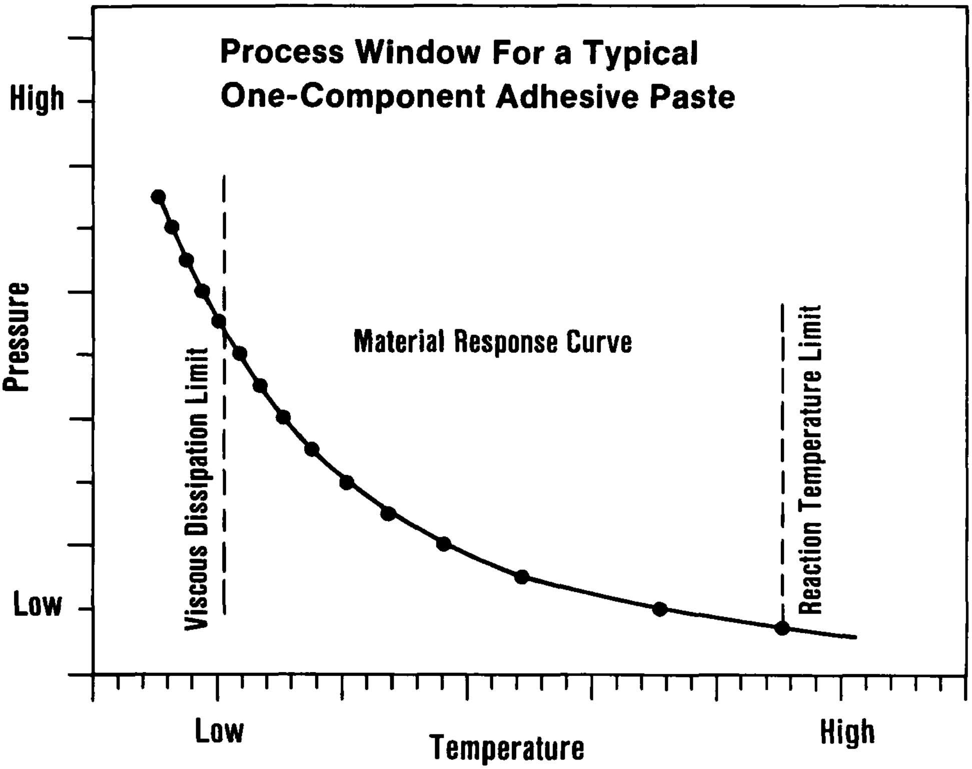

FIGURE 8 Process limits of a one-component adhesive.

flow-induced chemical instability will not be an issue for the adhesive dispensing process. The pressure, temperature, and/or flow geometry that makes the Brinkman number less than or equal to 1.0 defines the process limits. Figure 8 shows the process limit for a generic one-component adhesive in terms of pressure and temperature when a constant process output is desired. The temperature/pressure curve in the figure shows the material's response to the dispensing system. A low-temperature/high-pressure limit is set to avoid severe viscous-heating induced instability while a high temperature limit is due to the onset of the chemical reaction in the material. The adhesive paste is processable between the two limits.

SUMMARY

The process of dispensing one-component adhesives in automotive manufacturing applications was investigated in order to understand current processes and to guide new process development. The one-component adhesives, which exhibit non-Newtonian rheological behavior, were modeled as Bingham fluids possessing a yield stress and a steady-state viscosity. The quantitative model for the dispensing process includes four major flow sections, each described by a specific fluid-mechanics equation representing a component of the process hardware, connected in a serial configuration. The fluid-mechanics equations derived for Bingham fluids in the individual flow sections were solved simultaneously by numerical methods in order to understand the inter-relationships between the material variables (*e.g.* yield stress, viscosity, temperature dependencies) and process variables (*e.g.* pressure, flow geometry, temperature, output). The process model is generic and can be modified for any forced-flow adhesive application process. Important conclusions from the modeling study are:

1. Newtonian fluid mechanics does not adequately describe the dispensing process due to the apparent yield phenomenon of the adhesives. The flow profile (velocity distribution) changes from a parabolic shape for a Newtonian fluid to a "plug" shape for a Bingham fluid.
2. Dispensing temperature, among the process variables, has the strongest effect on the process output. For example, a ± 1 °C temperature perturbation can cause as much as a 14% variation in size of the adhesive bead dispensed, provided other variables are held constant.
3. It was also shown that each adhesive material, due to its flow characteristics, differs in processibility and non-linear sensitivity between pressure and temperature or valve opening and temperature. The non-linear sensitivity can be eliminated by operating the dispensing process isothermally.
4. Viscous heating, if it becomes important in the dispensing process, is likely to occur in high-shear-rate regions such as near the wall of the connecting pipe or in the metering valve.
5. The relative importance of flow-induced chemical instability for an adhesive dispensed at a set of process conditions can be calculated by introducing a redefined Brinkman number that takes into account viscous dissipation, heat conduction and convection during processing, and the difference between processing temperature and the onset temperature of chemical reaction.
6. Finally, the process limits imposed by the chemical stability of one-component adhesives can be defined by choosing process conditions such that the Brinkman number is less or equal to unity.

NOTATION

C_p	heat capacity
D	dimension (for example, diameter) where viscous dissipation is important in a flow
E_η	activation energy of viscosity
E_σ	activation energy of apparent yield stress
k	thermal conductivity
l_b	length of the adhesive bead
l_i	length of the i th flow segment
P	dispensing system pressure
ΔP_i	pressure drop in the i th flow segment
q	flow rate of the dispensing system
q_i	flow rate of the i th segment of the flow path
R_i	radius of the flow path

S_v	rate of viscous dissipation
t_c	dispensing process cycle time
T	dispensing temperature
T_r	on-set temperature: temperature at which chemical reaction rate becomes important on the time scale of the dispensing process
v	axial velocity
V	average axial velocity
V_z	velocity profile (distribution) in axial direction
α	empirical constant for measuring the temperature dependence of the dispensing system
γ_w	near-wall shear rate
η	viscosity
ρ	density
σ_0	apparent yield stress

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