Die Swell of Carbonaceous Mixes

by

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Introduction

In the manufacturing of carbon and graphite artifacts, the main purposes of the extrusion process are to maximize the density of green artifacts and to form green artifacts with a size and shape as near as possible to the end product.[1,2] However, the expansion of a green artifact upon leaving the die, the die swell, not only reduces its density, but also increases the amount of material to be machined off in the finishing process. Thus, it is desirable first to understand the causes of the die swell and then to better control the die swell phenomenon. It is known in the field of polymer processing that die swell is a result of the non-Newtonian (or viscoelastic) characteristics of the polymer at processing conditions. However, the increases in extrudate diameter in polymer processing (i.e., die swell) can be as high as one hundred percent (100%),[3,4] which are two orders of magnitude higher than the ca. one percent (1%) increase in electrode diameter usually noted in the carbon artifact manufacturing. Furthermore, coal tar pitches are generally characterized as Newtonian fluids at the extrusion temperature.[5,6] Hence, it is very likely that it does not take a sophisticated viscoelastic model to explain the die swell in the forming operation. This paper describes a simple model in which the die swell phenomenon is accounted for by the relaxation of gas pressure within the closed interparticle porosity.

Theory

A schematic diagram of a plunger-type extruder along with the closed porosities in a carbonaceous material within the extruder and the green electrode is shown in Figure 1. Assuming that: (i) the coke-pitch mix proper is incompressible, and (ii) the entrapped gases, including air and any pitch volatiles, follow the ideal gas law, two simultaneous equations can be established to describe the system shown in Figure 1:

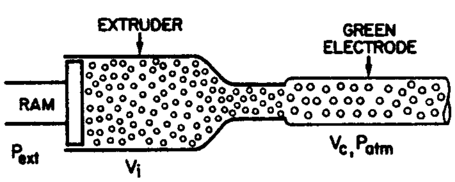

Figure 1. A Schematic Diagram of the Die Swell Model.

$$V_i\, P_{ext} = V_c\, P_{atm} \tag{1}$$

$$\frac{V_c + V_{c+p} + V_o}{V_i + V_{c+p} + V_o} = (1 + \text{rad. D.S.})^2 \times (1 + \text{long. D.S.}) \tag{2}$$

where:

- P_{ext} = the extrusion pressure experienced by the coke-pitch mix in the mud cylinder,
- P_{atm} = atmospheric pressure,
- V_i = closed porosity in the extruder, which is filled with compressed gases,
- V_c = closed porosity in the green electrode,
- V_o = open porosity, which should remain constant during the forming operation,
- V_{c+p} = the volume of coke-pitch mix proper,
- rad.D.S. = radial die swell ($\equiv \Delta D/D$), and
- long.D.S. = longitudinal die swell ($\equiv \Delta L/L$).

By substitutions and rearrangements, Equation (3) can be derived from these equations.

$$\frac{V_c}{V_g} = \frac{A - 1}{A\,(1 - B)} \tag{3}$$

where:

- A = $(1 + \text{rad. D.S.})^2 \times (1 + \text{long. D.S.})$,
- B = P_{atm}/P_{ext}, and
- V_g = the volume of a green electrode. $\sim V_c + V_{c+p} + V_o$.

Since V_g, P_{ext}, rad. D.S., and long. D.S. can be measured in the laboratory, the amount of closed porosity, V_c, required to account for the die swell can be calculated using Equation (3). This calculated closed porosity, V_c, is then compared with various types of measured closed porosity in the green electrode. If the proposed model is valid, the calculated V_c should compare favorably with the measured closed porosity.

Experimental

Coke-pitch mixes were prepared from a petroleum coke filler and a coal tar binder pitch with a Mettler softening point of ca. 110°C. The binder content was about 23.0-25.5 weight percent. Extrusions were performed using a laboratory 19 mm extruder. Porosity distributions in green electrodes were determined from various density measurements on the raw materials, mixes, and extruded electrodes.

The diameter of the extruded electrodes was measured using a micrometer. The radial die swell was then calculated from the diameter of the die, 19.15 mm, and the measured electrode diameter. The longitudinal die swell was determined by accurately measuring the travel of the ram and the extruded electrodes. Both the speed and distance of ram travel were first amplified by a rack-and-pinion assembly with an amplification factor of ten. The amplified ram travel was then measured by an electronic rate transducer (BEI Electronics, Model H25D). The extrusion rate and total travel of green electrodes were gauged by another rate transducer. On the basis of these data, the longitudinal die swell can then be calculated using the following equation:

$$\frac{V_{elec} \cdot D^2_{elec}}{V_{ram} \cdot D^2_{mud}} = \frac{L_{elec} \cdot D^2_{elec}}{L_{ram} \cdot D^2_{mud}} = (1 + \text{rad. D.S.})^2 \times (1 + \text{long. D.S.}) \quad (4)$$

where V's are the speed at which the ram or the extruded electrode travels; L's are the total distance of the ram or electrode travel; D_{elec} is the measured electrode diameter; and D_{mud} is the diameter of the mud cylinder, 76.25 mm.

Results and Discussion

The closed porosities in green electrodes can be classified into three groups. They are the intraparticle porosity, the closed interparticle porosity, and the total closed porosity which is the sum of the first two groups. The correlations between the calculated closed porosity [i.e., the V_c/V_g from Equation (3)] and these three types of measured closed porosities are shown in Figures 2-4. In addition, a diagonal line indicating a one-to-one correspondence between the calculated and measured closed porosities is also shown in Figures 2-4. The majority of the data points should fall on this diagonal line or in the vicinity of it, if the die swell is indeed caused by the expansion of gases entrapped in the measured closed porosity of interest. As can be seen in Figures 2 and 3, both the total closed porosity and the intraparticle porosity measurements are distributed below the diagonal line, indicating that the calculated closed porosity is mostly less than these two measured closed porosities. For the closed interparticle porosity (Figure 4), the measured porosity data are generally scattered around the diagonal line. In view of the difficulties associated with the longitudinal die swell and porosity distribution measurements, the extent of data scattering in Figure 4 is by no means excessive. Hence, it can be concluded that, to a large extent, the die swell phenomenon is a consequence

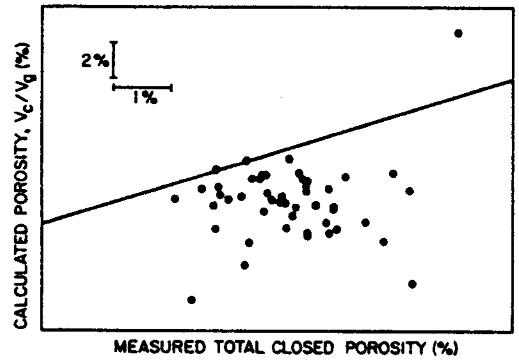

Figure 2. The Correlation Between the Calculated Closed Porosity and the Measured Total Closed Porosity.

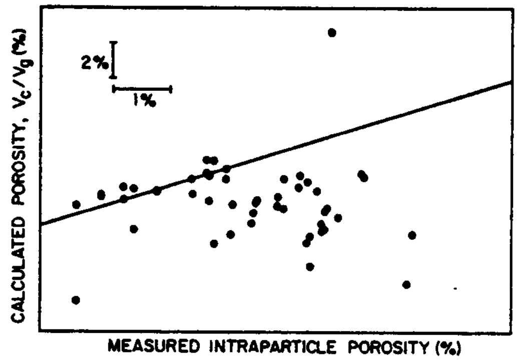

Figure 3. The Correlation Between the Calculated Closed Porosity and the Measured Intraparticle Porosity.

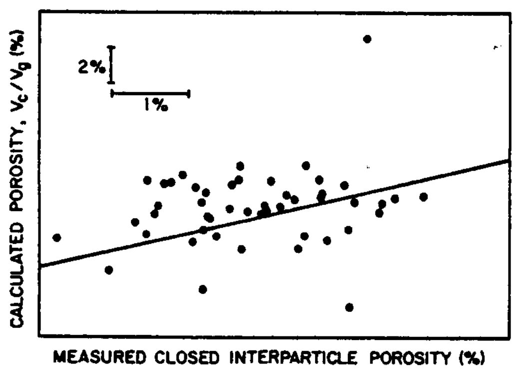

Figure 4. The Correlation Between the Calculated Closed Porosity and the Measured Closed Interparticle Porosity.

of the relaxation of gas pressures within the closed interparticle porosity in green electrodes.

References

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