VISCOSITY TESTING OF PITCHES SHEAR RATE AND TIME DEPENDENCE

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Introduction

The rheological behavior of pitches is very important in the production of graphite electrodes used on electric arc furnaces, since coal tar pitch is used as a binder and petroleum pitch is used as an impregnant. The standard characterization tests for pitch such as softening point, quinoline insolubles, toluene insolubles and coking value provide useful but limited information.

It is the purpose of this study to more fully characterize the rheological properties of coal tar and petroleum pitches in the context of the equipment and procedures that could be employed by an industrial quality control or testing laboratory.

Experimental

Viscosity was measured for the pitches listed in Table 1 using a Brookfield Thermosel system which consisted of a LVTD rotational viscometer and an electrically heated thermocontainer capable of temperatures to 300°C. For all testing, an SC4-31 spindle was used inside a cylindrical sample holder for a coaxial cylinder geometry. Sample volume was 10 ml.

The analog outputs for temperature and viscosity were acquired on a datalogger at 10 second intervals for later analysis.

Because of the strong effect of temperature on viscosity, both the pitch sample and spindle were preheated for 30 minutes before the spindle was lowered into the sample. In addition, 30 more minutes were allowed for temperature equilibrium before the spindle was rotated.

For testing at different shear rates, rotational speeds of 3, 6 and 12 rpm were used. These corresponded to nominal shear rates of 1.02, 2.04 and 4.08 sec^{-1}. Higher speeds generally resulted in off-scale readings at the normal test temperature of 160°C. Temperature was controlled to within 0.1°C.

Results and Discussion

It has been reported that fluids with high fine particulate content, such as paints and printing inks, show thixotropic behavior[1]. Although coal tar binder pitches may be considered to contain "fine particulate" in the form of QI, little work has been done to investigate possible thixotropic behavior. To explore this area, coal tar pitches A through F, with softening points ranging from 108.4°C to 112.3°C, were tested for viscosity vs time at 160°C and a nominal shear rate of 4.08 sec^{-1}.

The results are displayed in Figure 1 and show that viscosity decreased with time for all pitches, indicating thixotropic behavior.

Thixotropy is ascribed to the coal tar binder pitches in Figure 1 based on the simple straightforward definition of thixotropy. However, thixotropy can also be determined by viscosity response to a cycle of increasing and then decreasing shear rate. Lower viscosity during the decreasing shear rate portion is further evidence of thixotropic behavior. Figure 2 shows the response of pitch E to such a cycle and indeed apparent viscosity was lower as the shear rate was reduced back to the initial value.

Additional viscosity vs time testing was carried out on 3 laboratory prepared coal tar pitches from Aristech (now Koppers) to determine the effect of QI level. Figure 3 shows that pitch G with 15.6% QI showed a viscosity decrease with time as seen in Figure 1, while pitches

H and I with 10.1% and 10.2% showed very little decrease. This is further reinforced by reference to Figure 4 which shows that Ashland 240 (pitch J) with essentially no QI and very little beta resin had essentially no viscosity change with time and very little change with increasing shear rate.

Conclusions

Viscosity measurements using a rotational viscometer at 160°C with shear rates of 1-4 sec^{-1} have shown that coal tar pitches with 15% QI exhibit thixotropic behavior. This behavior is a function of QI content since thixotropy is minimal for coal tar pitches with 10% QI and absent for a petroleum pitch with essentially no QI.

Use of viscosity testing as a quality control tool for coal tar pitches is not straightforward since any measured apparent viscosity value may be a function of sample shearing history, test shear rate and shearing time.

Table 1. Characteristics of Pitches

ID	Type	SP °C	QI Wt%	TI Wt%	Beta Resins Wt%	Notes
A	CTP	112.3	16.2	28.5	12.3	Commercial
B	CTP	112.0	15.9	28.5	12.6	Commercial
C	CTP	110.7	15.9	28.9	13.0	Commercial
D	CTP	110.6	15.9	27.5	11.6	Commercial
E	CTP	109.7	15.5	27.4	11.9	Commercial
F	CTP	108.4	15.9	27.7	11.8	Commercial
G	CTP	110.3	15.6	27.6	12.0	Lab Prep
H	CTP	109.5	10.1	23.3	13.2	Lab Prep
I	CTP	112.0	10.2	23.3	13.1	Lab Prep
J	PP	121.8	Nil	3.9	3.9	A240

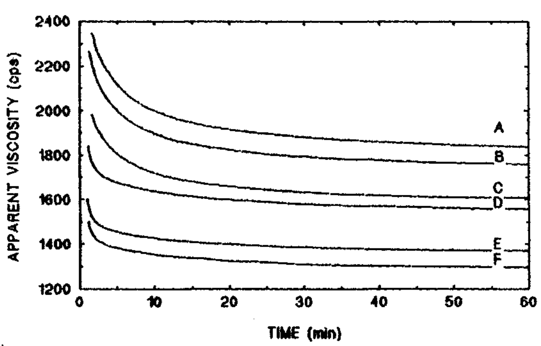

Figure 1. Apparent Viscosity vs Time for CT Pitches A through F at 160°C and 4.08 sec^{-1}

Acknowledgment

I would like to thank Ken Krupinski of Koppers Inc. for coal tar pitches G, H, and I prepared in his laboratory.

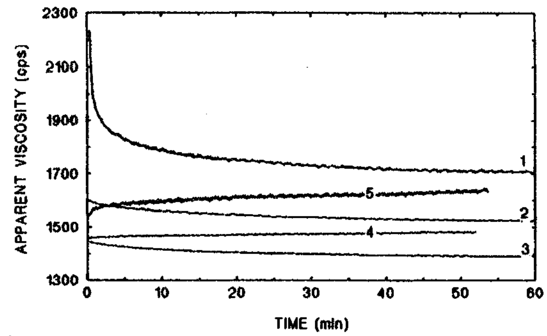

Figure 2. Apparent Viscosity vs Time for CT pitch E at 160°C in shear rate order of 1: 1.02 sec^{-1}, 2: 2.04 sec^{-1}, 3: 4.08 sec^{-1}, 4: 2.04 sec^{-1} and 5: 1.02 sec^{-1}

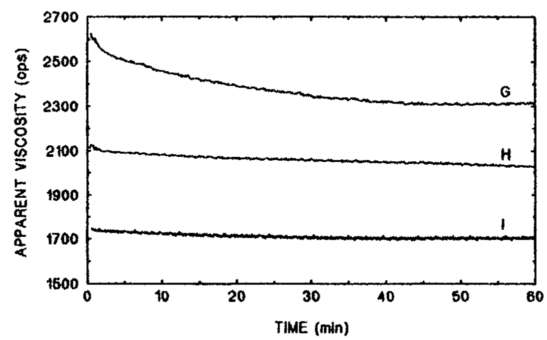

Figure 3. Apparent Viscosity vs Time for CT pitches G, H and I at 160°C and 1.04 sec^{-1}

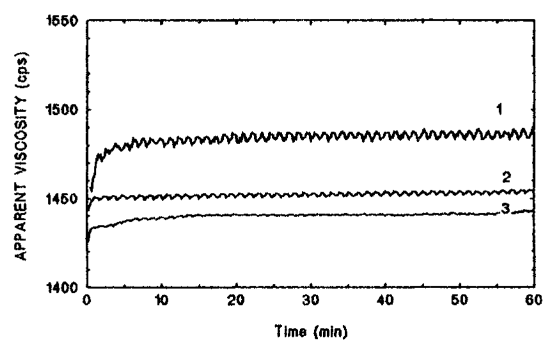

Figure 4. Apparent Viscosity vs Time for Petroleum pitch J (A240) at 165°C in shear rate order of 1: 1.02 sec^{-1}, 2: 2.04 sec^{-1} and 3: 4.08 sec^{-1}

Reference

1. Pierce, P.E., *J Paint Technol.*, 1971, **43**, 35.