INFLUENCE OF ADDITIVES ON THE VISCOSITY OF COAL TAR PITCH AND PITCH/FILLER-MIXTURES FOR SØDERBERG ELECTRODES

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INTRODUCTION

A trouble free operation of Søderberg-electrodes requires a low viscous green paste. For a given amount and composition of filler material the viscosity of the green paste is determined by the choice of the binder. Coal tar pitches with higher softening points can be used after plasticizing by adding low boiling reflux oils. Model compounds which can be found in reflux oils were used to study the plasticizing effect on coal tar pitch simply by adding them to the pitch and analyzing the resulting softening point and viscosity. Based on these results a model for the plasticizing of coal tar pitch was developed, which could successfully be used to find even more effective additives. Four commercially available compounds were added to a binder pitch for production of green paste. These green pastes were tested with respect to their viscosity.

EXPERIMENTAL

The coal tar pitch was heated up to 140°C, the compound added in an amount of 5%, and that mixture stirred for 5 min with 500 rpm. The viscosity was determined with a capillary viscometer. The capillary was made of stainless steel; it was 1,29 mm in diameter and had a length of 50 mm; the viscometer volume was 250 ml and the pitch was forced through the capillary by an argon pressure. The viscosity was calculated by the Hagen-Poiseuille equation [1]. Green pastes were produced in a 10 l sigma-blade-mixer. The given filler composition was preheated and mixed with the binder for 20 min at a temperature of 150°C. Samples of 320 g were compacted with 5 bar to a cylinder of 75 mm in diameter and appr. 45 mm in height. The viscosity of the paste was determined at a temperature of 100°C with the Elkem-prototype compression-viscometer [2].

RESULTS AND DISCUSSION

Figs. 1(a) and 1(b) show the logarithm of viscosity in dependence of the reciprocal absolute temperature for a coal tar pitch to which different model compounds were added. The stronger temperature dependence of the viscosity at lower temperatures is assumed to be caused by the dissolution of preordered molecular structures, like e.g. larger molecule aggregates [3]. Additives have the strongest effect in that region. The results shown in Fig. 1(a) indicate that the viscosity of the coal tar pitch is stronger reduced if the basicity of the functional group of the additive is increased. In a more general approach for commercial additives it was found that small, pitch-compatible Lewis bases are very effective plasticizers. Four of the most effective additives (A,B,C,D) reduced the viscosity of the pitch by nearly the same extent (see Fig. 1(b)) and even more effective than 1-naphthylamine. The results lead to the following idea: Coal tar pitch, especially at lower temperatures, contains large aggregates of molecules which are responsible for the high viscosity and their strong temperature dependence. Such aggregates can be nucleated by two large pitch molecules, one being a Lewis base and the other a Lewis acid. Once being formed, the nucleus can attract further pitch molecules. Additives being small molecules but strong Lewis bases are able to replace the Lewis bases of the pitch with the consequence that the aggregate is destroyed; the viscosity is reduced to lower values. The four most effective additives reduced the viscosity of the coal tar pitch studied nearly by the same extent but the influence on the viscosity of the green pastes was quite different; see Table 1. This can be understood by different selective adsorption of the additives at the filler particle surfaces and thus differently reduced attractive forces between the filler particle surfaces and large pitch molecules being Lewis bases.

CONCLUSIONS

A model for plasticizing of coal tar pitch with Lewis bases was developed. A similar reduction of the viscosity caused by these Lewis bases in the pure coal tar pitch does not necessarily mean a similar effect in mixtures with filler material. In the latter case the additive effect is determined by the interaction with the filler particle surfaces.

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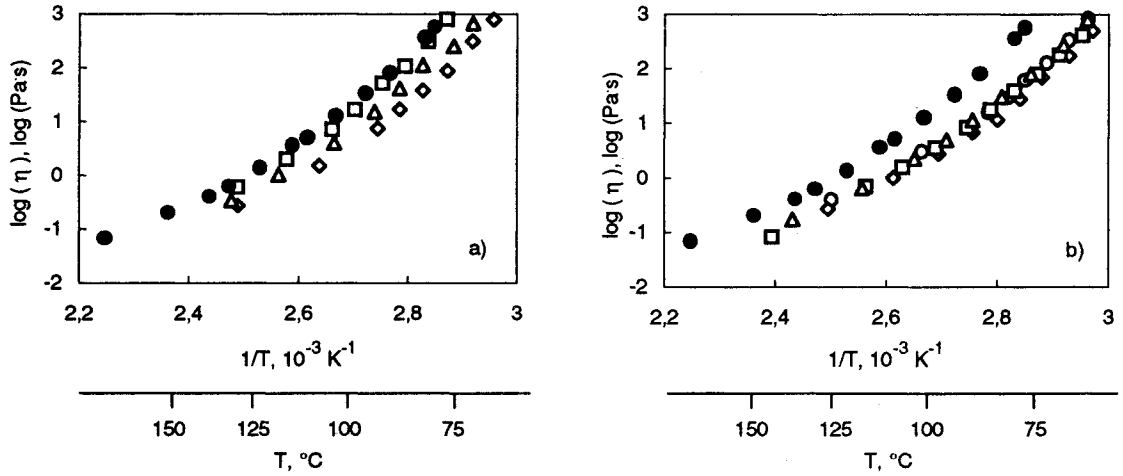

Figure 1: Logarithm of viscosity in dependence of the temperature; additive content in CTP: 5%
a) ● original CTP,□ CTP + 1-naphthoic acid,△ CTP + 1-naphthol,◇ CTP + 1-naphthylamine
b) ● original CTP,△ CTP + A,◇ CTP + B,○ CTP + C, □ CTP + D; A,B,C,D = Lewis bases

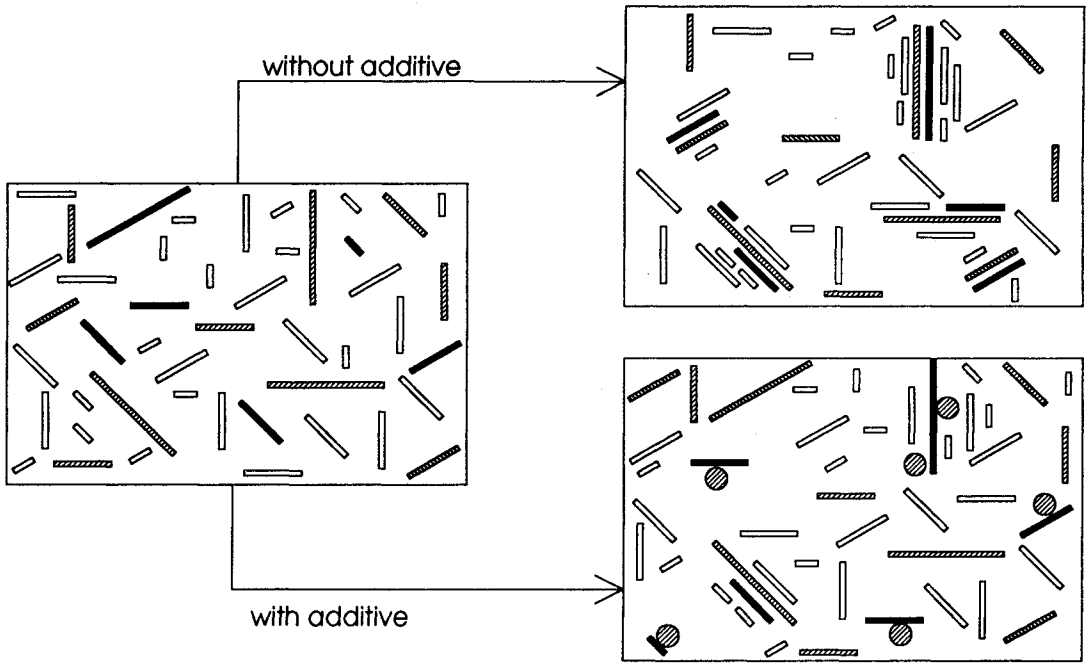

Figure 2: Model of pitch in hot liquid state (left side) and cooled state with aggregates (right side)

Neutral compounds, Lewis acids, Lewis bases of the pitch; molecules of different sizes

Additive molecule, Lewis base

Table 1: Viscosity of pastes with identical filler material, binder and additive content but different additives; measured at T=100°C, speed of compression=3 mm·min^{-1}; results in kPa·s.

Original CTP	CTP + A	CTP + B	CTP + C	CTP + D
1880	1090	1320	1510	1830