Abstract:

Laser Desorption Ionization Mass Spectrometry was applied for analysis of coal tar pitch and its thermal reaction. Pitch was found to consist of polycyclic aromatic hydrocarbons without alkyl side chains, ranging from m/z)*=160 to 6 000. Although compounds with molecular weights m/z=500 or more were observed only in TI (toluene insoluble) in the pitch, compounds below m/z=500 were found in both TS (toluene soluble) and TI. Heat treatment increased the peak intensity of the TI u*

ble) or QI (quinoline insoluble) is often used as an

important index to describe its composition, which relates to properties such as carbonization yield.

Instrumental analysis has also been applied to pitch in order to characterize its chemical structure. Nuclear magnetic resonance spectroscopy and infrared spectroscopy are used to determine the average structure of pitch[1]. The chemical constituents in pitch are analyzed by using gas chromatography and liquid chromatography, despite the analytical limitation of molecular weight[1,2]. Size exclusion chromatography and mass spectrometry are efftro Ş Ş ĸ el,6)

, field desorption[7,8], laser desorption ionization (LDI)[2,3], and matrix assisted laser desorption ionization[2,3] are applied for analysis of constituents with high molecular weights. Since ionization methods influence the degree of fragmentation and detection sensitivity[9], the most suitable ionization method is chosen for the purpose of characterization.

In this report, LDI mass spectrometry was chosen to find out the chemical structure and thermal reaction scheme of pitch constituents, because it is suitable for analysis of solid samples.

Pitch A was obtained by distillation of coal tar which produced by a commercial coke oven (operated at JFE Steel Corporation). The primary QI was removed before distillation. 2.0 kg of the QI-free tar was distilled in a glass flask at 320˚C under flowing nitrogen (flow rate: 1.5 L/min) with a pressure of 31 kPa. The resulting Pitch A has a softening point (SP) of 77˚C.

*) Mass to charge ratio

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[*ghi*] perylene (276) to naphto [1, 2, 3, 4−*ghi*] perylene (326)[8].

In order to observe the high molecular weight constituents, both TS and TI were examined by LDI mass spectrometry, since peaks of high molecular weight ions were not found in the spectrum of the whole Pitch A. The spectra are shown in for TS and in (a) for TI.

In comparison with the whole pitch (see Fig. 1), the TS shows a very similar spectrum to the pitch, while the TI shows a wider molecular distribution, which ranges from *m*/*z*=167 to around 5 000 or 6 000. Figure 3 (b) presents the mass spectra of TI in a narrow *m*/*z* range for an easy comparison between TS and TI. As in the TS, constituents having *m*/*z* from 167 to 500 were found. Whereas TS shows the highest intensity at around *m*/

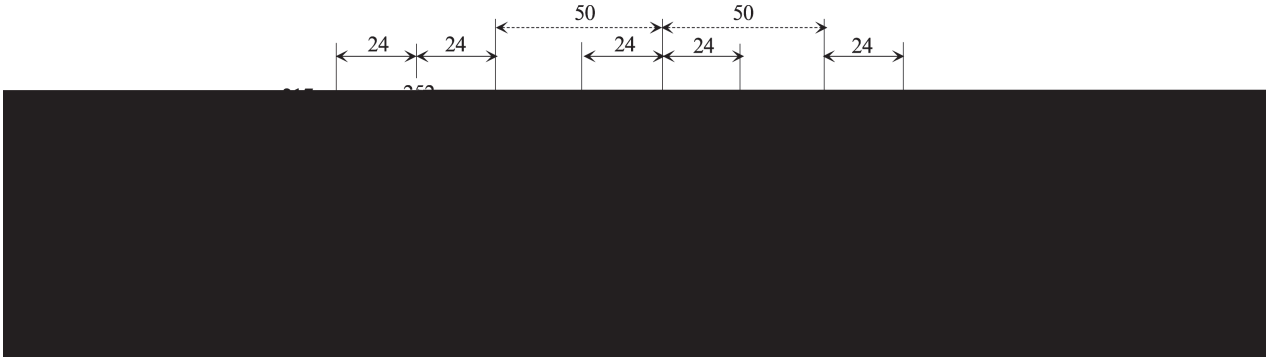

Fig. 1 LDI Mass spectrum of pitch A

Fig. 2 LDI Mass spectrum of TS in pitch A

Fig. 3 LDI Mass spectrum of TI in pitch A

pitch by the liquid mixture, and the excess amount over its solubility will precipitate as TI[10].

The same constituents were found in both TS and TI in the spectrum ranging from *m*/*z*=167 to 500 (see Fig. 2 and Fig. 3 (b)). These constituents are considered to exist in pitch in a larger concentration than its solubility, and for this reason, the constituents will be found both in TS and in TI. Since molecular weight is one of the major determining factors of solubility, the constituents of high molecular weight with *m*/*z*=500 or more are found only in TI. However, the amounts of some constituents such as *m*/*z*=256, 270, 306 are larger in TI than in TS, which means that the solubility will be affected not only by the molecular weight but also by other factors such as polarity.

In the heat treatment of pitch, dehydrogenation of polycyclic aromatic hydrocarbons causes the poly-condensation reaction, and the molecular weight of the constituents in the pitch becomes higher than before heat treatment. This reaction converts TS constituents to TI, and as a result of this conversion, the amount of TI increases. As described in the results section, there was almost no difference in the molecular weight distribution in the TS spectra before and after heat treatment (see Fig. 2 and Fig. 5). This fact implies that the specific

spectra, and the reaction schemes were also discussed.

JFE Chemical Corporation has been producing pitch products such as binders and impregnation pitches by a production process developed over the course of many years. We will continue to produce products which meet customers' needs by carrying out basic analysis and developing production technologies.

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