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Stepwise preparation of petroleum-based impregnation pitch for synthetic graphite

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Introduction

Synthetic graphite is utilized in various fields such as electrodes for electrical arc furnace for steelmaking, crucibles for semiconductors manufacturing, carbon brushes for electric motors, and etc. In the synthetic graphite production, the impregnation pitch (IP) plays an important role to enhance the properties such as density, strength, modulus, and etc. The two most important properties required for impregnation pitches are low softening points and high coking values, which are very difficult to achieve simultaneously, due to their trade-off relationship and inherent complexity. We introduce a stepwise preparation technique to synthesize impregnation pitches from pyrolysis fuel oil (PFO), a residue from petroleum refinery. The properties of experimental IPs obtained from the stepwise technique are investigated, and the IPs were examined to a lab-scale impregnation process to ensure impregnation capabilities.

Materials and Methods

The impregnation pitch was prepared from PFO using the stepwise preparation technique. The first step, the pressurized heat treatment, was carried out at 370-430 °C for 3 hours in the autoclave reactor, and the system pressure increased naturally with the gas generated during heating. The second step, the atmospheric heat treatment, was carried out at 390 °C for 3 h under atmospheric pressure. The IPs were labelled P37A, P39A, P41A, and P43A, depending on the temperature of the first step, respectively. The pitch prepared by the atmospheric heat treatment (one-step preparation) without the first step was named as AT. Carbon bodies were prepared to simulate the impregnation process, and the impregnation capabilities of the experimental IPs were evaluated by measuring density enhancement and porosity reduction.

Results and Discussion

Figure 1 shows the softening point, the coking value, and product yield of the IPs. Whereas softening point of AT was 132 °C, those of the IPs, prepared by stepwise technique, decreased below 130 °C. The stepwise technique induced the cracking and the polymerization reaction simultaneously. Furthermore, the process suppresses the removal of light compounds and forces them to participate in the polymerization reaction[1-2]. As a result, the softening point increase was suppressed, and the coking values were increased from 47% up to 54%. In particular, the product yield increased up to 45%, which is more than 10% higher than that of the one-step treatment of 32%. Among the experimental pitches, P39A possess the softening point of 118 °C and the coking value of 49%, which were comparable with the commercial one.

The carbon bodies, which have the density of 1.40 g/cm^3 and the porosity of 29%, were impregnated and re-carbonized using the experimental pitches. Impregnation performances of all the IPs obtained by stepwise preparation were superior to that of AT. The IPs increased the density of the carbon bodies up to 1.60 g/cm^3 and thus the porosities decreased down to 18.6%. On the other hand, P41A and P43A did not improve the properties of the carbon body even though they had higher coking value than others. This was due to the presence of quinoline insoluble (QI), which prevent the pitch components travel through the channels in the body (or may have acted as an obstacle) during the impregnation process[3].

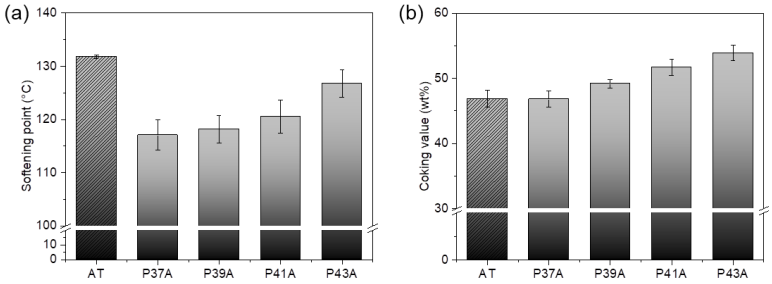

Figure 1. Characterization results of the impregnating pitches prepared by stepwise technique: (a) softening point (b) coking value.

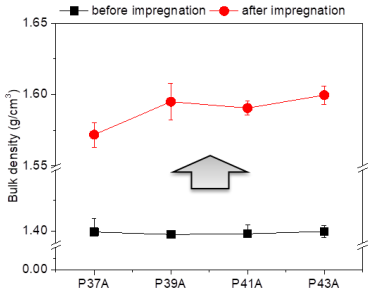

Figure 2. Bulk density of the carbon body after applying to impregnation

Conclusions

The stepwise preparation technique was proven to be effective to synthesize the impregnating pitch from PFO. The technique was capable of enhancing the pitch yield and the coking value of the pitches, and of control the softening point simultaneously by inducing the polymerization of light compounds. Moreover, it confirmed that the formation of QI can disrupt the impregnation capabilities.

Acknowledgment

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References

1. J.-E. Choi, S. Ko, J.G. Kim, Y.-P. Jeon. (2018). Effect of pressure during the synthesis of petroleum pitch precursors in open and closed system. *Carbon letters* 25, 95-102
2. M. Martínez-Escandell, P. Torregrosa, H. Marsh, F. Rodríguez-Reinoso, R. Santamaría-Ramírez, C. Gómez-De-Salazar, E. Romero-Palazón. (1999). Pyrolysis of petroleum residues: I. Yields and product analyses. *Carbon* 37, 1567-1582
3. J.-E. Choi, S. Ko, Y.-P. Jeon. (2019). Preparation of petroleum impregnating pitches from pyrolysis fuel oil using two-step heat treatments. *Carbon Letters*, in press