# DEVELOPMENT OF HYPER COAL DERIVED CARBON FIBER

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## Introduction

For the development of electric vehicles (EVs) and plug-in type hybrid electric vehicles (PHEVs), the weight reduction of automobile is one of most important tasks to improve the energy efficiency to improve the lack of power and a short driving range in the conventional EVs. Carbon fiber reinforced plastic (CFRP) which has lower weight and higher strength than steel is considered as a suitable alternative as car-body material of such automobiles. Jim deVries of the Ford Motor Company recommended that the tensile strength, elongation ratio, and Young's modulus of CF for car frames be at least 1.7 GPa, 1.5%, and 170 GPa, respectively, with a material price less than 10 USD/kg[1]. Polyacrylonitrile-based CFs (PANCFs) exhibit a higher tensile strength, elongation ratio, and Young's modulus than the recommended mechanical properties. However, its production cost is more than 20 USD/kg due to expensive polyacrylonitrile precursor fiber and its low carbonization yield. On the other hand, mesophase pitch-based CFs (MPCFs) have high mechanical properties comparable to PANCFs, but the applications of MPCFs are very limited because they have the high production cost due to low yield of spinnable mesophase pitch. For example, the yield of mesophase pitch derived from decant oil by hydrogenation using 1, 2, 3, 4-tetrahydroquinoline and heat treatment is 5.0–10.0 wt%[2]. Hyper-coal (HPC) is one of the cheap resources for high performance functional carbons, which is prepared by direct solvent extraction of coal using methylnaphthalene at 350–400$^{o}$C under high pressure[3]. It has very interesting characteristics of low ash, high carbonization yield, and excellent thermoplastic properties. In this work, we prepared spinnable mesophase pitch with high pitch yield through the usual three-step process of hydrogenation, $N_2$ blowing heat treatment, and TLE using HPC as the raw material.

## Materials and Methods

HPC and 1, 2, 3, 4-tetrahydronaphthalene (tetralin) were mixed at 1:1 and 1:2 ratios (w/w) and heat-treated at 400–450°C for 1–4 h under autogenous pressure using an autoclave. After removing tetralin, the samples were heat-treated at 415°C for 3–4 h with $N_2$ blowing heat treatment, and light molecular components were removed by TLE at 390°C for 10 min under vacuum. The molecule structure and molecular weight distribution of obtained pitches were estimated by $^{13}$C solid-state nuclear magnetic resonance spectroscopy ($^{13}$C-NMR) and time-of-flight mass spectrometry (TOF-MS). After producing the pitch fibers by a single-hole spinneret with a laboratory-type mono-hole melt spinning apparatus, obtained fibers were stabilized (270°C, 0.5°C/min, Air 200 mL/min), carbonized (1000°C, 20°C/min, 30 min, vacuum) and graphitized (2,800°C, 15°C/min, 10 min, Ar atmosphere). The structure of the transverse sections and the surface morphology of the graphitized fibers were observed using a scanning electron microscope (SEM). The mechanical properties of the carbonized and graphitized fibers were measured using a tensile tester in accordance with the JIS R 7606:2000 method.

## Results and Discussion

The results of TOF-MS and $^{13}$C-NMR of the obtained pitches exhibited the hydrogenation of HPC decreased the amount of methylene chains and heavy molecular components with high polynuclear aromatic compounds. The $N_2$ blowing heat treatment was necessary to reveal the mesophase texture but not to increase the molecular weight and mesogens, including aromatic carbons. The obtained mesophase pitch showed many bulk flow textures with less than 20% isotropic spheres by volume (**Figure 1. (a)**). The mesophase pitch showed very high pitch yield of 54.9 wt% and softening point of 267°C. The striation in the fiber axis direction was observed on the surface of the obtained fiber, and the radial-random structure was observed on the cross-section of the obtained fiber (**Figure 1 (b)**). **Table 1** summarizes the mechanical properties of the HPC-derived carbonized fiber. The tensile strength, elongation, and Young's modulus of the carbonized fiber were 1.8 GPa, 1.4%, and 140 GPa, respectively. The tensile strength of the carbonized fibers was high enough to meet the objective CFs.

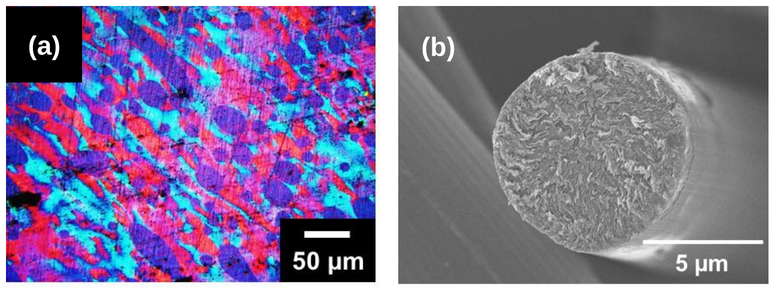


**Figure 1. (a) Anisotropic structure of HPC-derived mesophase pitch and (b) SEM image of HPC-derived MPCF after graphitization.**

**Table 1. Mechanical properties pf HPC derived carbonized fiber**

| | Stabilization | Carbonization | | | | |
|---|---|---|---|---|---|---|
| | Yield | Yield | Diameter | Tensile strength | Young's modulus | Elongation |
| | [wt%] | [wt%] | [μm] | [GPa] | [GPa] | [%] |
| HPC-derived MPCF | 109.0 | 84.7 | 10.2±0.5 | 1.8±0.4 | 140±20 | 1.4±0.3 |
| Target CFs | 108–110 | >85 | 8–10 | 1.7 | 170 | 1.5 |

## Conclusions

Spinnable mesophase pitch at an enormous yield of 54.9 wt% was successfully prepared through the three-step manufacturing process of hydrogenation, $N_2$ blowing heat treatment, and short TLE using HPC as an effective raw material. HPC-derived MPCF showed high tensile strength of 1.8 GPa and modulus of 140 GPa after carbonization.

## Acknowledgment

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