PRODUCTION OF MESOPHASE FROM A COAL TAR PITCH IN A SEMI-PILOT POLYMERIZATION REACTOR

A.T. Gontijo - Centro de Pesq. e Desenv. da USIMINAS, Ipatinga, MG, Brazil, CEP. 35160-900
C. Otani - Dept. de Física do ITA-CTA, S.J.Campos, SP, Brazil, CEP.12228-901
S. Otani - Div. de Materiais do IAE-CTA, S.J.Campos, SP, Brazil, CEP.12228-904
C. A. Luengo - IFGW, DFA, GCA, UNICAMP, Campinas, SP, Brazil, CEP.13083-970

Introduction

Anisotropic phase formation and growth, in an isotropic pitch media, are associated with complex physico-chemical processes which are intermediate steps of the pyrolitic transformation towards carbon materials [1-2]. These reactions are strongly dependent on process parameters and also on the chemical characteristics of pitches used as raw materials [2-3].

An experimental device was designed and constructed to establish better conditions for mesophase formation in a coal tar soft pitch, produced by the USIMINAS metallurgical coke plant.

Experimental

To perform the experimental work, an isotropic soft pitch produced by USIMINAS was selected, whose main characteristics are presented in Table I [4]. The reactor properly constructed for this objective is schematically presented in Figure 1.

Table I. Main characteristics of USIMINAS soft pitch.

	SP °C	QI %	TI %	Ash %	FC %	H/C %
Soft Pitch	40	3.5	12.1	0.1	35.2	64.7

SP = softening point, QI = quinoline insoluble fraction, TI = toluene insoluble fraction, Ash = ash content, FC = fixed carbon and H/C = ratio of elementary hydrogen and carbon contents.

The experimental apparatus has the following main components : a) steel made 5.0 liters process chamber, 15.5 cm inner diameter and 30.0 cm high; b) mechanical stirring system with four blades distributed in two levels, rotation of 380 rpm induced by an electric motor; c) annular nitrogen injection system with four equidistant holes, positioned at intermediate depth between two sets of stirring blades; d) volatile material exhausting system at top of the reactor, coupled with condenser and liquid/gas separation system plugged to a vacuum pump; e) three thermocouples at different levels and radii to measure the temperature profile inside the reactor; f) electrical furnace, its temperature is measured and controlled by an external thermocouple linked to a microprocessed PID system; g) manually operated valve at the bottom of the reactor which can be used to discharge the sample; h) liquid sample collector and temperature lowering chambers.

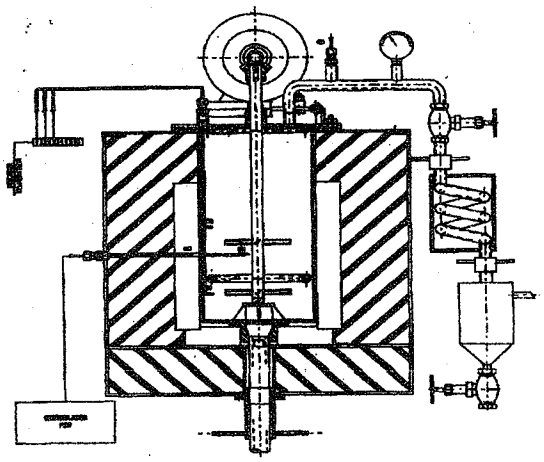

Figure 1. Schematic diagram of semi-pilot polymerization equipment.

Using the semi-pilot polymerization reactor, the low-softening-temperature pitch samples were heat treated at temperatures varying from 420 to 520°C, under the following conditions: stirring 380 rpm, nitrogen flux 20 Nl/min, initial charge 3,500 ml, initial temperature : 100°C, heating rate of furnace : 100 °C/min.

The sampling interval for each test temperature was fixed to 1.0 h, for all isotherm durations.

Results and Discussion

The heat treated soft pitch samples were analyzed to obtain their softening points, quinoline insoluble and toluene insoluble fractions. The structural analysis was done by determination of anisotropy or mesophase content (Aniso) by optical microscopy. The time dependence of SP temperature and QI, TI and Aniso contents are graphically presented in Figure 2.

The representative optical micrographs obtained by analysis of the samples heat treated at different temperatures are presented in Figure 3.

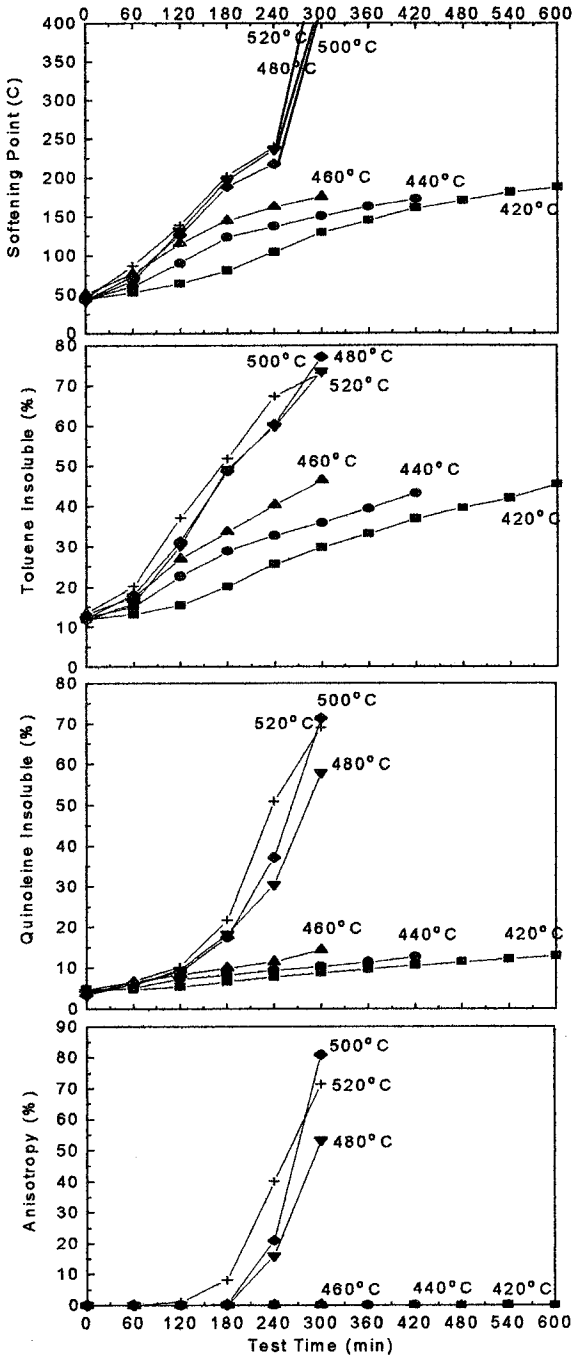

Figure 2. Time dependence of SP, TI, QI and Aniso contents for treatment temperatures indicated in graphs.

The correlated analysis of all characterization results of heat treated soft pitch samples indicated that : a) there is a limiting time t >3 h and temperature T > 460°C for which a rapid increase in SP temperature, QI and Aniso contents is observed; b) the TI contents gradually increase with increase in the time interval and temperature range tested; c) the long time treatments at temperatures lower than 460°C can produce pitch with sparsely distributed small size (less than 4 μm) mesophase spheres, and these treatments provide pitches with high TI (~45%) and low QI(~15%) contents; d) the number and average size of mesophase spheres grow as a function of heat treatment temperature increase, and the maximum value of Aniso, about 80%, is achieved at 500°C, i.e., for thermosetting pitch.

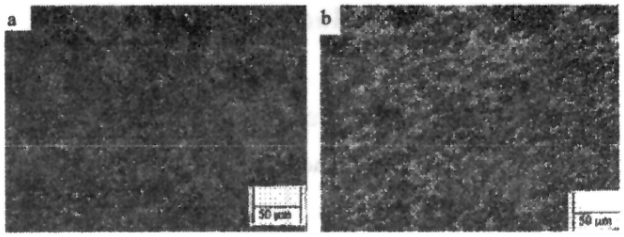

Figure 3. Photomicrography of soft pitch heat treated at (a) 480 °C, and (b) 500 °C.

Conclusions

The experimental results yield the following conclusions : a) the semi pilot polymerization set up is useful for mesophase production parameter studies; b) the heat treatment of soft pitch at temperatures close to 500°C is suitable for mesophase production; c) the soft pitch without pre treatments can be partially converted to mesophase (~80%) but its thermoplasticity will be lost as a consequence of two phenomena simultaneously occurring in the pitch : the growth of the mesophase and the polymerization of the isotropic pitch compound.

Acknowledgment

We are grateful to USIMINAS who provided the samples and equipment and also for FAPESP processo 94/0644-2 for financial support.

References

1. Marsh, H. and Menendez, R., in Introduction to Carbon Science, Marsh, H., Chap. 2 : 37-33, Butterworths, 1989.
2. Bermerjo, J. et alii., in Fuel, 74 (12) : 1792-1799, 1995.
3. Otani, S.; Otani, C.; Polidoro, H.A. and Weyne, G.S., in, Int. Symp. on Carbon, Tsukuba, Japão p 758, 1990.
4. Gontijo, A.T., Otani, C., Otani, S. and Luengo, C.A., "The study of QI extraction from coal tar pitch using coal tar derived oils as solvent in a centrifugation process", presented in this conference.