

# WHY CARBONS OF DIFFERENT STRUCTURES ARE DEPOSITED IN CHEMICAL VAPOR INFILTRATION OF POROUS STRUCTURES

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## Introduction

In chemical vapor deposition of carbon from light hydrocarbons, carbons with different structures may be obtained by changing the deposition conditions; they are termed as rough laminar (RL), smooth laminar (SL) and isotropic (ISO) [1]. The same structures are obtained in chemical vapour infiltration (i) by changing the infiltration conditions or (ii) with progressive infiltration of a porous structure at constant infiltration conditions [2]. The present paper is concerned with a model to explain the formation of different structures with progressive infiltration. It is based on deposition chemistry and kinetics, pore surface area/pore volume ratio and in particular on the interaction of deposition chemistry and kinetics with the surface area/volume ratio. Due to its fundamental character the model is also applicable to explain formation of different structures by changing infiltration conditions.

## Experimental

A carbon fiber felt, 20 mm in thickness, with a fiber volume fraction of 12 % was infiltrated at 1100 °C and pressures of 20 kPa (pure methane, methane-hydrogen mixture) and 30 kPa (methane-hydrogen mixture). The mixture exhibited a partial pressure ratio of 7 : 1. Infiltration occurred in the absence of pore diffusion limitations. The mass gain of the felt as a function of infiltration time was determined. The structure of the infiltrated carbon was analysed by polarized light microscopy. Studies by high resolution SEM and TEM are presented in a further paper of the conference [3].

## Results

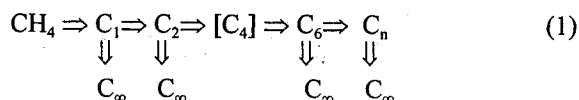
The mass increases of the felt as a function of infiltration time demonstrate overall infiltration rates increasing with increasing methane partial pressure. Maximum infiltration

rates exist at the beginning of infiltration. This observation is most important for derivation of the model. In this stage the felt exhibits the lowest internal surface area, which implies that surface related deposition rates are much higher in this stage than in subsequent stages of infiltration.

A typical polarized light micrograph of an internal cross-section of a felt after infiltration is shown in Fig. 2. According to the optical activity three layers may be detected: (1) A first layer with very low optical activity. (2) A second layer with low optical activity and high preferred orientation (see extinction contours). (3) A third layer with high optical activity and low preferred orientation. These layers correspond to (1) ISO, (2) SL and (3) RL carbon.

## Derivation of the Model

Carbon deposition from methane is based on the interaction of complex gas phase and surface reactions [4].



$\text{C}_1, \text{C}_2, \text{C}_4 = \text{C}_1, \text{C}_2, \text{C}_4$  - hydrocarbon species;  $\text{C}_6, \text{C}_n$  = mono- and polynuclear aromatic hydrocarbon species;  $\text{C}_\infty$  = solid carbon.

The deposition rates increase with increasing number of carbon atoms of a hydrocarbon species. At a low surface area/volume ratio gas phase reactions are favored, i. e.  $\text{C}_6$  and  $\text{C}_n$ -species are rapidly formed and surface related deposition rates are high. At a high ratio surface reactions are favored and carbon will mainly be deposited from species with a low number of carbon atoms such as  $\text{C}_2$ -species, which show much lower surface related deposition rates. These opposite conditions exist at the beginning and the end of infiltration, respectively.

A corresponding model is presented in Fig. 3. It demonstrates that infiltration rates decreasing with increasing degree of infiltration can only be correlated with an increase of the surface area/ volume ratio. At the beginning of infiltration the internal surface area increases until the layers growing around the fibers lead to a fiber bridging (first stage of infiltration). As the pore volume is simultaneously decreased, the surface area/volume ratio is increased. With progressive infiltration the internal surface area is decreased (second stage of infiltration), but the volume decrease is stronger, i. e. the surface area/volume ratio is further increased. This consideration confirms a continuous increase of the surface area/volume ratio from the beginning to the end infiltration. It implies that carbon deposition is changed from polynuclear to mononuclear and light linear hydrocarbon species. Consequently, the deposition rates decrease and the structures of the carbon change from ISO to SL and RL

### References

- [1] Bokros JC. Deposition, structure and properties of pyrolytic carbon. In: Walker PL, editor. Chemistry and physics of carbon, Vol5, New York: Dekker, 1969:1-118.
- [2] Benzinger W, Hüttinger KJ. Chemistry and kinetics of chemical vapor infiltration of pyrocarbon-VI. Carbon 1999; 37: in press.
- [3] Reznik B, Gerthsen D, Hüttinger KJ. Structural studies of chemical vapor infiltrated pyrocarbon. This conference.

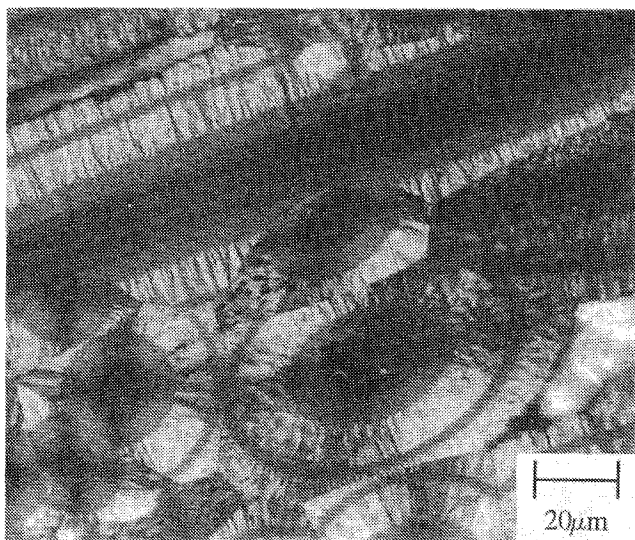


Fig.2 Polarized light optical micrograph of a cross-section of felt II.

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### Acknowledgement

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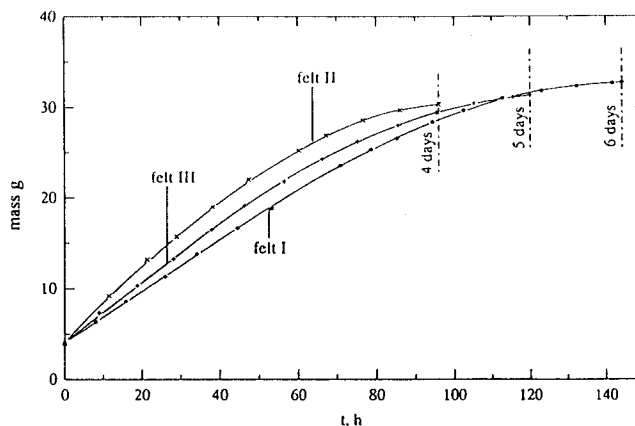


Fig.1 Mass increase of the felt as a function of infiltration time. (felt I) total pressure of 20 kPa,  $CH_4/H_2 = 7:1$ ; (felt II) total pressure of 30 kPa,  $CH_4/H_2 = 7:1$ ; (felt III) total pressure of 20 kPa, pure methane.

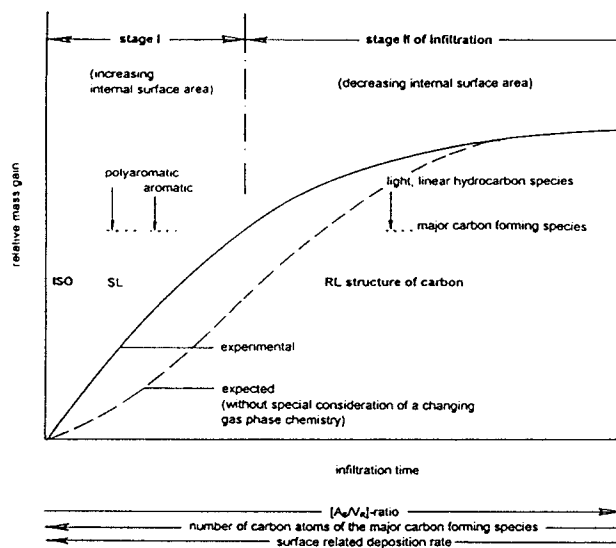


Fig.3 Model showing the formation of ISO, SL and RL carbon with progressive infiltration.