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Towards the preparation of pure graphite fluoride with $(C_2F)_n$ structural type

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Graphite fluorides (GF) are used in numerous fields, ranging from cathode electrode in lithium batteries, solid lubricant or for hydrophobic surfaces applications. They even have the potential to be part of a new generation of cold neutron reflector. GF have first been synthesized by Ruff and Bretschneider in 1934 [1], with a composition of $CF_{0.92}$. Then, a series of studies had given birth to poly(carbon monofluoride) $(CF)_n$ compounds. It is only fifty years later that poly(dicarbon monofluoride) $(C_2F)_n$ compounds were synthesized by Watanabe and his team [2-3]. They demonstrated that the fluorination temperature needed to be low in order to synthesize the $(C_2F)_n$ structure (Fig. 1a). On the other hand, high reaction temperature leads to the $(CF)_n$ phase.

$(C_2F)_n$ phase is particularly interesting because of its stacking sequence FCCF/FCCF where half of the carbon atoms are non-fluorinated but with sp^3 hybridization (FCF/FCF for $(CF)_n$ type). On one hand, the interlayer distance is the highest for the covalent F-Graphite Intercalation Compounds, *i.e.* 0.9 nm. On the other hand, $(C_2F)_n$ can be used as a precursor for the preparation of F-diamane via an exfoliation route; this phase can be described as a stacking of F-diamane layers along the *c*-axis.

Because $(C_2F)_n$ is an intermediate phase and the high reactivity of elemental fluorine gas (F_2), it is difficult to prepare this phase with high purity. Most of the GF with F/C ratio close to 0.5 reported in the literature are mixtures of $(C_2F)_n$ with graphitic or/and $(CF)_n$ phases. In our work, fluorination conditions were adapted with very long duration (72 h) and starting with graphite precursors with large granulometries (44 and around 200 μm).

In this study we present the different synthesis that led us to obtain nearly pure $(C_2F)_n$ phase from different graphite precursors (powders and foil) and at different temperature (Fig. 1b). To this extent, a structural characterization was done with XRD, NMR and TEM observations (Fig. 1c), while the C-F bonding was investigated by FTIR and NMR studies. In particular, ^{13}C NMR evidences the absence of sp^2 carbons and high relative content of diamond-like carbon (non-fluorinated sp^3 C typical of $(C_2F)_n$), *i.e.* 45% whereas the theoretical value is 50%.

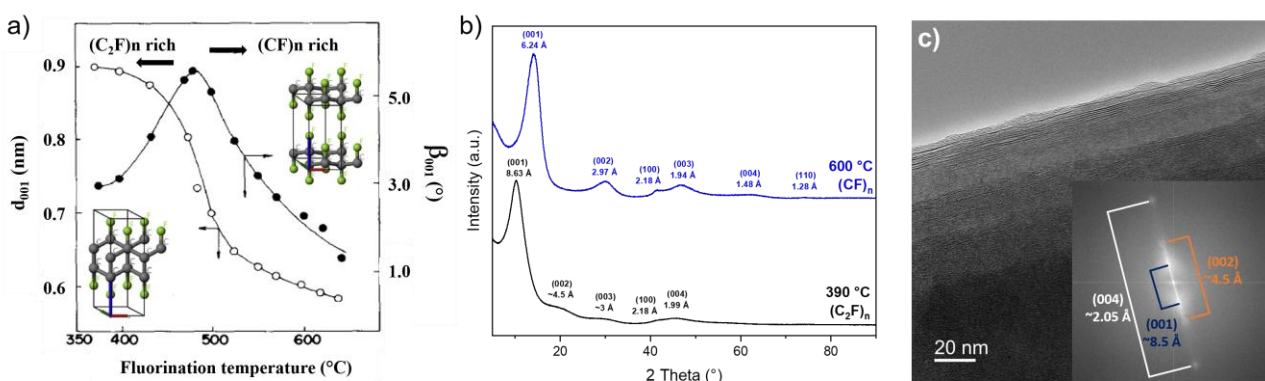


Fig. 1: a) Interlayer spacing (d_{001}) and half widths (β_{001}) of the (001) reflection of fluorinated graphites as a function of the fluorination temperature. Adapted from [2,3]. b) X-ray diffraction pattern (Cu $K\alpha$, 1.5406 Å) of a thick foil of graphite (1 mm) fluorinated at 390 and 600 °C. c) HRTEM image of powder of fluorinated graphite. The FFT image in the right corner confirm the $(C_2F)_n$ structure since d_{001} is around 0.85 nm.

References

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