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Origin of cometary and chondritic refractory organics: Ion irradiation experiments

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Abstract

The formation process of Refractory Organic matter present in chondrites and Interplanetary Dust Particles (IDPs) of cometary and asteroidal origin is a debated issue. Earlier studies have advocated a formation step in a hot environment, however the potential role of ion irradiation has been so far poorly constrained. We present here experimental simulations that address this issue, comprising thermal degradation and ion irradiation experiments conducted at GANIL (Caen France) and CSNSM (Orsay France). We show that unlike thermal reactions, ion irradiation might lead to ROM-like material under very stringent conditions on both the nuclear dose and the nature of precursor. These very narrow conditions suggest that forming ROM without any action of thermal reactions is extremely difficult in astrophysical environments, either ISM or the proto-solar disk.

1. Introduction

Refractory organic matter (ROM) – termed Insoluble Organic Matter (IOM) in chondrites – is a polyaromatic carbonaceous solid ubiquitous in cometary dust and primitive chondrites. The origin of this material is still a debated issue. While the high D/H fractionation points to a formation step in low-T conditions, the polyaromatic structure might require an energetic input as thermal processing and/or ions irradiation [1-4]. We report here experimental simulations in order to test both these processes for producing kerogen-like materials from various precursors: polyethylene glycol (PEG1450), sucrose, lignine, cellulose and a Me-OH soluble extract of lignite.

2. Results and Discussion

Thermal degradation experiment were run at IPAG with a tubular furnace maintained under secondary vaccum, over the range 300-1000 °C. Low-energy (LE-) irradiations were performed on the IRMA beamline at CSNSM (Orsay-France) with fluences up to 4.10^{14} ions/cm^2 [C 40 keV, Ne 170 keV], and high-energy (HE-)irradiation at GANIL (Caen-France) [Zn 590 MeV, C 12 MeV, Ni 12 MeV]. Infrared spectra were collected *in situ* during irradiation. Raman spectroscopy (514 and 244 nm) was performed *ex situ* to characterize the polyaromatic structure of the samples. Both IR and Raman data allowed comparison with IOM/ROM from chondrites/cometary dust. Heating experiments resulted in the carbonization of the insoluble precursors lignine, cellulose and sucrose. PEG 1450 fully sublimated during warming, and no data were collected from the lignite extract. The carbonization process resulted in carbon enrichment (H and O loss), along with aromatization and growth of aromatic units. Samples obtained at 400°C during ~1h were found fairly similar to chondritic IOM, showing similar functional groups and polyaromatic structures. Nevertheless, they were not strictly similar and a weak, but significant precursor effect was observed. We also suspect that kinetic effects were at play. In contrast, LE-irradiations failed in providing relevant analogs. All precursors were transformed into an almost amorphous carbonaceous material above a critical nuclear dose of ~ 10 eV/atome. No progressive structural transition was observed. HE-irradiation provided a more complex picture. Irradiated PEG1450 and lignite-extract led to insoluble dark solids whose composition was controlled by the electronic dose. They showed a global composition similar to IOM, but also critical differences (e.g. acetylinic group C≡C, which is absent in IOM) (Fig. 1, 2). In addition, these samples were fluorescent and fragile, precluding the

collection of Raman data. We infer their sp^2 structure was very dissimilar from a polyaromatic structure. In contrast, HE-irradiation of lignine and sucrose led to polyaromatic samples, but more disordered than that of IOM. This precursor effect is consistent with previous studies on polyimide and soots [5], suggesting that the final structure is strongly controlled by the structure of the precursor.

3. Conclusion

These results show that heating processes provide fair ROM-like polyaromatic solids, and kinetics and precursor effects might even improve these analogs. In contrast, LE-irradiations lead to amorphous carbonaceous material, structurally dissimilar to ROM. HE-irradiation is a plausible chemical route to transform simple species into an insoluble solid, but its respective structure and composition would be dissimilar from ROM. Producing a ROM material would require highly aromatic precursor, but even in this case fitting ROM composition appears very difficult. Finally, our experiments support the view that thermal processing was likely a step of the formation process of ROM.

References

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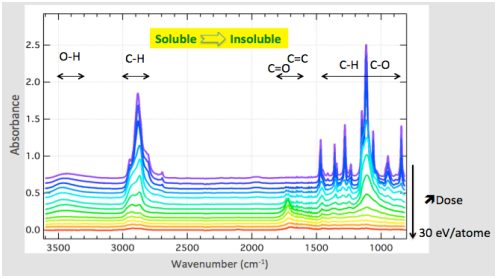

Figure 1: Infrared spectra of PEG1450 during a high energy irradiation (electronic regime).

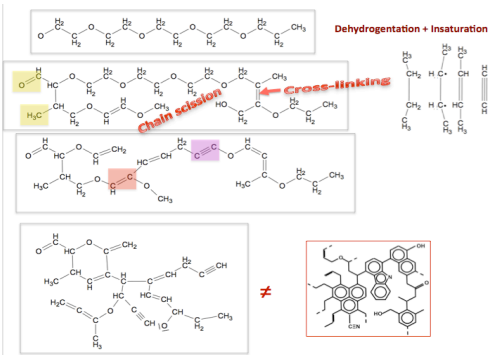

Figure 2: A schematic view of the structural and chemical evolution of the PEG1450 polymer under high-energy irradiation. A macromolecular insoluble material is finally formed, but it is dissimilar to a polyaromatic structure (in red box).