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PRODUCTION OF HIGH MOLECULAR WEIGHT ORGANIC COMPOUNDS ON THE SURFACES OF AMORPHOUS IRON SILICATE CATALYSTS: IMPLICATIONS FOR ORGANIC SYNTHESIS IN THE SOLAR NEBULA I. Gilmour¹, H. G. M. Hill², V.K. Pearson¹, M.A. Sephton¹ and J. A. Nuth². ¹Planetary and Space Sciences Research Institute, The Open University, Milton Keynes MK7 6AA, U.K. ²Code 691, NASA Goddard Space Flight Center, Greenbelt MD 20771, U.S.A. (i.gilmour@open.ac.uk, hill@lepvax.gsfc.nasa.gov)

Introduction: The Fischer-Tropsch-type (FTT) and Haber-Bosch (HB) syntheses are elegant and efficient mechanisms for producing organic matter from simple precursors. The requirements are the simple combination of CO, H₂ (plus N₂ for HB synthesis) and a mineral catalyst at temperatures of several hundred °C. For this reason, catalytic synthesis was widely considered as the most probable means of producing organic matter in the early Solar System [1].

Previous work on the catalytic synthesis of organic matter in the 1960's and 1970's [e.g., 1] employed a variety of catalytic materials including Fe-Ni meteorites, Fe particles or powdered carbonaceous meteorites. However, these materials are not appropriate analogues for the types of catalysts that may have been available in the primitive nebula [2]. Subsequent work on the relatively uncontaminated meteorites such as Murchison and isotopic studies of the proposed meteoritic FTT products, has cast doubts over an FTT synthesis for meteoritic organic matter. Several key compounds previously attributed to this mechanism were shown to be terrestrial contaminants [3]. Given the abundance of hydrogen and CO in the primitive solar nebula, however, it is important that the potential for the FTT synthesis of organic matter is fully investigated.

Silicate grain formation in O-rich circumstellar outflows starts with SiO nucleation followed by rapid condensation of additional refractory elements to form highly-reduced amorphous grains [4]. Analogs of these "astronomical silicates" are made at Goddard Space Flight Center. The amorphous iron silicate smokes are produced by the combustion of vapors (Fe-SiO-H₂-O₂) and the overall composition of the grains cluster around the metastable eutectics, greenalite, and iron-rich saponite dehydroxalate.

The Fe silicate smokes have remarkable catalytic properties [2, 5]. We constructed a closed gas circulation system whereby basic synthesis gases (CO, N₂, H₂) were continuously passed through ~0.5 g of Fe silicate smoke at temperatures of 200-600°C and pressures close to 1 atm. Using continuous FTIR spectroscopy, the gas was found to become increasingly enriched with key prebiotic molecules (methane and its homologs, water, ammonia, and CN-bonded compounds) via Fischer-Tropsch (FT) and Haber-Bosch (HB) syntheses. Here we report the results of an ex-

periment designed to identify and quantify chemical species produced and remaining on the surface of the smokes used for an extended series of FT/HB syntheses. We compare these species with the organic matter found in carbonaceous meteorites.

Organic Analysis of Smokes: The organic matter associated with two smoke samples and one blank sample were analyzed using pyrolysis-gas chromatography mass spectrometry. The samples (ca. 20 mg of powdered catalyst) were flash pyrolysed at 610°C followed by analysis on an Agilent Technologies 5973 GCMS [6].

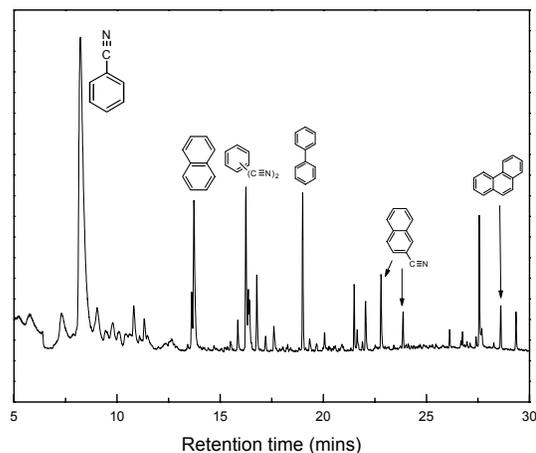


Figure 1. Total ion current chromatogram of pyrolysate released from the surface of an Fe-silicate catalyst used for 9 consecutive FTT and HB-type runs in the temperature range 200-600°C.

Results: Two smoke samples were examined in this study. The first (FESIL91500) was processed in the presence of CO (150 Torr) and hydrogen (~650 Torr), the second (FESIL102700) was processed in the same manner but half the CO in the original mixture was replaced with nitrogen.

Smoke FESIL91500 gave mean abundances of 7.9 wt% C and <0.02 wt% N. FESIL102700 gave mean abundances of 11.6 wt% C and 0.16 wt% N.

The total ion current chromatogram for sample FESIL102700 is shown in Fig. 1. The amount and complexity of organic matter released on pyrolysis was greater for sample FESIL102700 which is dominated

by a strong peak with a retention time of around 8.24 mins. and identified as benzonitrile from its mass spectrum. Other aromatic compounds are also present and abundant including benzene, toluene, naphthalene, methyl- and cyano-substituted benzenes and biphenyl. In general aromatic and substituted aromatic species predominate over aliphatic species. FESIL91500 also yielded aromatic species including benzene and toluene, however, yields were low and only trace quantities of compounds such as benzonitrile and naphthalenes could be detected. A more detailed examination of the products released from the catalyst surfaces by pyrolysis can be undertaken by examining ions characteristic of species of interest and producing reconstructed ion chromatograms (RIC) that shows the distribution of these species. RICs for aromatic and aliphatic series of compounds are shown in Fig. 2.

Aliphatic Hydrocarbons: Figures 2a and 2b are m/z 57 RICs that selectively display the distribution of aliphatic hydrocarbons. An homologous series of n-alkanes was observed ranging from n-C₁₀ to n-C₁₉, with a maximum at n-C₁₄. However, aromatics predominate over this molecular weight range.

Nitriles and Naphthalenes: Figures 2c and 2d are RICs (m/z 128, 142, 156) that selectively displays the distribution of C₀-C₂ alkyl substituted naphthalenes and di-cyano benzenes. In addition to the significant amount of benzonitrile observed in the TIC (Fig. 1) for the catalyst exposed to N₂, it is apparent that significant quantities of nitrogen-containing aromatics are

synthesized.

Discussion: The results obtained in these experiments show both similarities and some marked differences to the results of previous FTT synthesis experiments. In their original studies, Anders and his colleagues [1] used the apparent isomeric selectivity of FTT, in particular a strong predominance of n-alkanes, to argue that such compounds in meteorites were formed by a catalytic synthesis in the early solar nebula. It is now commonly believed that the majority of straight chain hydrocarbons in meteorites represent terrestrial contaminants [3], however, there is clear isotopic evidence that aromatic species are indigenous to carbonaceous chondrites [6]. The data reported here, indicate that aromatic and N-containing aromatic species can be readily formed by FTT/HB syntheses on Fe-silicate smokes raising the possibility of a nebular origin for at least some of the compounds in meteorites.

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