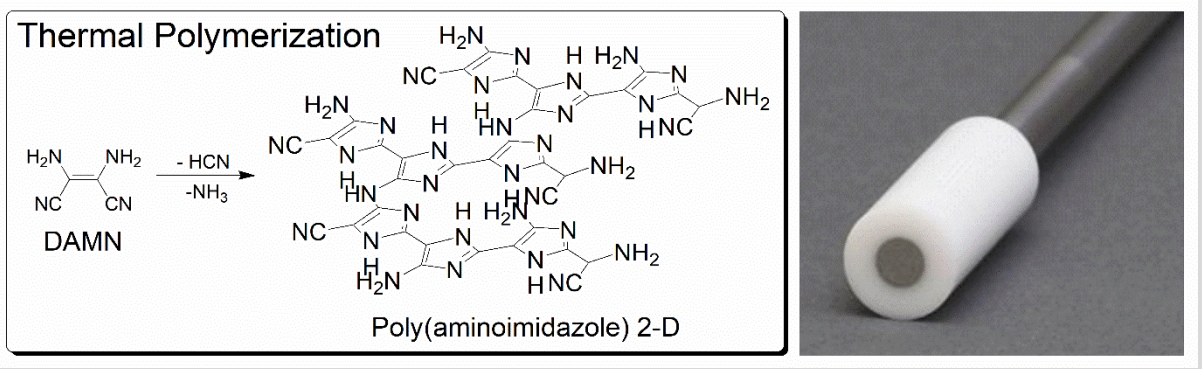
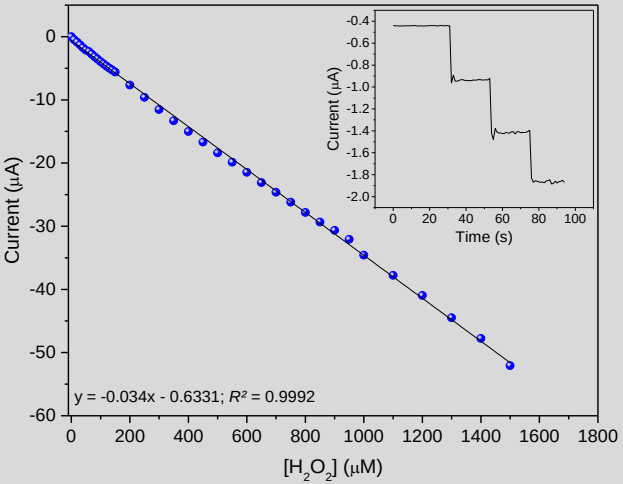
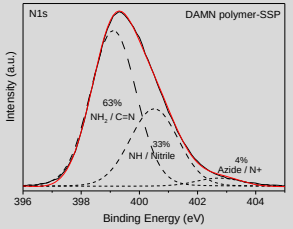
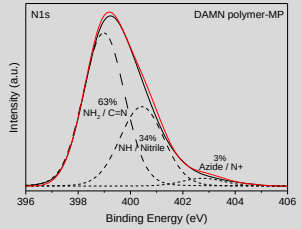
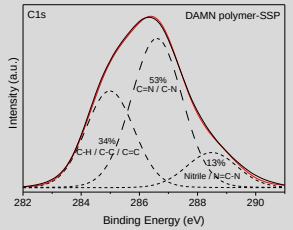
HCN-derived polymers from thermally induced polymerization of diaminomaleonitrile: A non-enzymatic peroxide sensor based on prebiotic chemistry

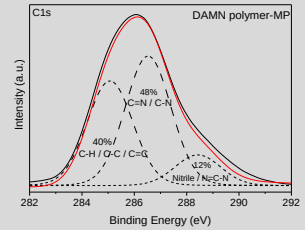












HCN-derived polymers have recently attracted considerable attention due to their promising applications as multifunctional materials. This study, inspired by plausible early Earth geochemical conditions, describes a strategy to synthesize them from the self-initiated thermal bulk polymerization of the HCN tetramer, diaminomaleonitrile (DAMN), with outstanding sensing properties. These conjugated polymers were obtained through non catalysed and simple isothermal reactions at 170 ◦C in the solid-state, and experiments at 190 ◦C permitted polymerization in the melt. Both processes are highly efficient, allowing quantitative yields of the end products. The conductivity properties of both polymers have been explored to show their high potential, especially DAMN polymers synthesized in melt, as non-enzymatic peroxide sensors. To better understand the differences found between the two series, structural characterisation was carried out using compositional data, Fourier transform infrared (FTIR), nuclear magnetic resonance (NMR) and X-ray photoelectron (XPS) spectroscopies, and X-ray diffraction (XRD) measurements. The interpretation of the structural data suggests that a two dimensional (2-D) macrostructure based on N-heterocyclics is predominant regardless of the state of monomer aggregation during the course of polymerization, but preferably formed in the melt. The morphological and thermal stability properties of the polymers based on DAMN were also evaluated. Finally, the most likely mechanisms based on the dehydrocyanation and deamination reactions that take place during the polymerization reaction are proposed. This study demonstrates the robust and straightforward character of these thermally activated polymerizations, which are of interest to chemical evolution research and to current materials and surface science.

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