

Solid state matter and chemical evolution in space. Insights from quantum chemical calculations

Albert Rimola
Departament de Química, Universitat Autònoma de Barcelona, 08193 Bellaterra, Catalonia, Spain (albert.rimola@uab.cat)

Abstract

Our universe is molecularly rich [1-3], comprising from the most essential molecules (such as H_2 , H_2O and NH_3), passing through complex organic molecules (namely, COMs, molecules between 6 – 13 atoms where at least one is C rendering its organic character [4]), reaching more evolved complex organic compounds, in which in some cases are of biological relevance, such as amino acids, nucleobases and sugars. Formation of early Solar-type systems involves successive steps, mainly represented by the prestellar, protostellar, protoplanetary disk, and planetesimal and planet formation phases [5]. This evolution goes hand-in-hand with an increase of the molecular complexity, in which more complex molecules form at each step. A key role of this chemical evolution is played by cosmic solid state materials (CSSMs), as their surfaces provide catalytic sites that favor pivotal chemical reactions. For instance, in the prestellar phase, grains actively participate in several surface-induced reactions such as formation of CH_3OH by H-addition to CO [6], while in the protostellar phase, COMs can be formed on the surfaces of the ices during the collapse stage [7]. The current knowledge of the role of the CSSMs in the chemical evolution occurring in space is mostly based on spectroscopic observations, helped by laboratory experiments and astrochemical models. This combination has been fruitful to elucidate the chemical composition, broad structural features and chemical activity of CSSMs. However, they cannot provide atomic-scale information such as their detailed structure or the actual role in chemical reactions, a serious limitation to fully understand the fundamental steps leading to the chemical complexity in space. This limitation can be alleviated by adding a fourth level of investigation grounded on rigorous quantum mechanical methods. In this talk, a couple of examples demonstrating the relevance of these theoretical methods to obtain

quantitative atomic-scale information are presented. The first example concerns the formation of formamide (NH_2CHO) as a COM test case. COMs formed on the surfaces of grains are believed to occur via radical-radical coupling [8], in the particular case of formamide by the coupling of NH_2 with HCO . However, formation of NH_3+CO has been identified as a competitive path against formamide formation. Additionally, other mechanisms leading to formamide formation have also been identified, such as reaction between CN with H_2O molecules belonging to the ice mantles [9]. The second example is related to the interaction of a set of molecules belonging to different families of organic compounds identified in meteorites of the class of carbonaceous chondrites (CCs) with silicates. Quantum chemical calculations have allowed establishing an affinity scale of these compounds with the forsterite (Mg_2SiO_4) silicate, one of the most abundant material present in both the matrix and chondrules of CCs. The affinities, based on the calculated binding energies of the organic compounds with the silicate, are compared with their abundances found in different meteorites [10].

References

- [1] Kwok, S., A&AR, Vol. 24, 8, 2016.
- [2] Tielens, A.G.G.M., Rev. Modern Phys., Vol. 85, pp. 1021-1081, 2013.
- [3] van Dishoeck, E.F., Faraday Discuss., Vol. 168, pp. 9-47. 2014.
- [4] Herbst, E.; van Dishoeck, E.F., ARA&A, Vol. 47, pp. 427-480, 2009.
- [5] Caselli, P., Ceccarelli, C., A&AR, vol. 20, pp. 1-68, 2012.

[6] Hama, T., Watanabe, N., Chem. Rev., Vol. 113, pp. 8783-8839, 2013.

[7] Garrod R.T., Weaver S.L.W., Herbst E., ApJ, Vol. 682, 283, 2008.

[8] Öberg, K.I., Chem. Rev., Vol. 116, pp. 9631-9663, 2016.

[9] Rimola, A., Skouteris, D., Balucani, N., Ceccarelli, C., Enrique-Romero, J., Taquet, V., Ugliengo, P., Submitted to ACS Earth Space Chem., 2018.

[10] Rimola, A., Trigo-Rodríguez, J.M., Martins, Z., Phys. Chem. Chem. Phys., Vol. 19, 18217-18231. 2017.