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Astrochemistry: an open molecular science cloud approach to low temperature reactivity

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Abstract – The appropriate formulation of low temperature rate coefficients values for use in modelling astrochemical systems is a typical test bed for the implementation of Open Molecular Science Cloud management tools for distributed calculation (or downloading), validation and re-use of Molecular Science knowledge. Here the case of an abstraction reaction forming a rather stable intermediate of interest for astrochemical studies is considered.

1. MOSEX: a project for providing Molecular Open Science Enabled Cloud Services

At present our activity in Open Molecular Science Cloud (OMSC) is driven by the wish to create Molecular Science (MS) contents for the European Open Science Cloud (EOSC) portal. EOSC aims, in fact, at developing Open Science cloud based services for creating a specific pan-European structure that generalizes the openness of data produced, develops interoperability and data sharing across both disciplines and infrastructures and enlarges as well the user base to researchers and innovators from all disciplines. In particular, the members of the MS community are at present designing and developing the Molecular Open Science Enabled Cloud Services (MOSEX) [1] infrastructure for:
a) *producing/discovering and downloading* MS data for the open collaborative and fully re-usable databases and repositories like CHEMCONNECT (for combustion) [2-11], ioCheM-BD (for mechanics modelling and materials physics) [3-12] and QCArchive (for quantum chemistry data) [4-13];
b) *running/checking/correcting (where possible) or discarding and validating* data of service (a) through iterative cycles collaboratively undertaken by the members of the community;
c) *annotating/curating/preserving* data of service (b) for its more efficient re-use;
d) *evaluating the quality of services* (a), (b) and (c) provided by the members of the MS community using Quality of Service (QoS) techniques;
e) *offsetting debits through credits of the adopted Prosumer (Producer+Consumer) model* in which the community members are at the same time users (making debits for used services) and producers (gaining credits for produced services) of the cloud.

3. Validation of rate coefficients of the H + CH_2 abstraction process

For the calculation of rate coefficients the collaborative efforts of the members of the MS community have been devoted to:
α) the construction of the potential energy surface governing the process,
β) the evaluation of the rate coefficient of the reactive process,
by developing the related tools in the appropriate area of knowledge. In particular:
Block α is the most typical MS theoretical and computational knowledge area and several computational tools of different quality (depending on the complexity of the molecular systems involved) are available for the purpose of carrying out ab initio electronic structure calculations and the subsequent formulation of the resulting potential energy surface as a suitable functional form in the cloud environment; using the MOSEX procedure the MHG [5-21] Potential Energy Surface

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(PES) exoergic by 4.1 kcal/mol, with no barrier to reaction and having a well of 115.4 kcal/mol in the strong interaction region (corresponding to the methyl radical stable geometry) was adopted.
Block β is either a classical trajectory or a quantum mechanics (or a mixed quantum classical one) dynamical treatment of the nuclei equations performed for the different initial collision and internal energy values necessary for evaluating thermal rate coefficients in the cloud environment.
The case considered here is the H + CH_2 reaction that plays an important role in astro-chemistry. Accurate estimates of its rate coefficients are needed in order to ground the computational modelling of some astro-chemistry phenomena on validated data. In particular, for the modelling of the properties of the inter-stellar medium, temperature values as low as 10 K need to be considered. Using the MOSEX procedure quasiclassical trajectory (QCT) estimates of the rate coefficient were obtained using a cloud version of Venus96 [6-22] after incorporating the MGH PES into it. The computed QCT values of the rates agree within a factor of two with the experiments of Fulle and Hipper in their J. Chem. Phys. 1997, 106, 8691-8698.

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