

## Preface Response to Reviewer 2

To facilitate the understanding of our response to both the Reviewer 01 and the Reviewer 02, we first put in perspective negative-ion catalysis. The fundamental mechanism underlying atomic negative-ion catalysis was proposed by our group in the context of muon catalyzed nuclear fusion [<https://doi.org/10.1088/0953-4075/43/20/201001>; doi:10.1088/1742-6596/225/1/012002]. The mechanism involves anionic molecular complex formation in the transition state (TS), with the atomic negative-ion breaking the hydrogen bond strength. The mechanism has been demonstrated in the synthesis of H<sub>2</sub>O<sub>2</sub> from H<sub>2</sub>O catalyzed using the Au<sup>-</sup> and Pd<sup>-</sup> anions to understand the experiments of Hutchings and collaborators [<https://doi.org/10.1039/B705915A>; <https://doi.org/10.1039/B509542E>; DOI: 10.1126/science.aad5705], in the catalysis of light, intermediate and heavy water to the corresponding peroxides [<https://doi.org/10.1021/jp301861q>], in the oxidation of methane to methanol without the CO<sub>2</sub> emission [<https://doi.org/10.1007/s13404-012-0056-7>] and more recently, in the doubly-charged negative ions as novel tunable catalysts: graphene and fullerene molecules versus atomic metals [<https://doi.org/10.3390/ijms21186714>].

Response to 1: The authors state novelty of the work given

a) The introduction of "double charged negative ions" as the activating mechanism for each catalyst rather than pure chemical additive that enhances the reaction and avoids elementary steps by polarizing ALL electrons in the systems such that "the double charge" instantly breaks the bonds of the reactants thereby creating "direct paths" to the products as a consequence of bond reformation after the transition states have been achieved.

b) furthermore, the mechanism of "double-charge" ions utilizing potential catalysts presented in this work has been well established by the works James Tour wherefore application of electrostatic spark is most efficient in converting a plethora of organisms and compound directly to elemental or allotrope carbon. Given that the electrostatic charge "in our case double charge negative ions" can be generated via nanoscale probes and other standard experimental tools we are presenting that can be used for mobile approaches such as recent drone technology that have been recently developed.

Response to 2

We use the previously well -established H<sub>2</sub>S oxidation to SO<sub>2</sub> in order to demonstrate that multi-step as well as direct path reaction mechanisms can actually be tuned or tailored to the most efficient energy barrier height depending on the catalyst used. In other words, It has been previously documented that the application of "doubly charged negative ions" to catalysts is efficacious for tuning direct path and multi-step reactions.

Response to 3

As the focus of this work has to do with "Relative Barriers" of one catalyst versus another and comparison of barrier heights all geometrical optimizations initial, final, and subsequent transition states that are predicted by utilizing Local Density Approximation (LDA) utilizing dmol3 standard code package as referenced in the citings

Response to 4

The catalysts are well-established Carbon Allotropes of spherical, cylindrical, and planar geometry as well as planar MoS<sub>2</sub>. The article is a comparative study of the Transition State energies as affected by the introduction of double charged negative ions.

Response to 5

Minimum basis sets file of 3.5 with Hexadecapole multipolar expansion via a self-consistent field (SCF) value  
.01Ha and .1 Ha smearing value

#### Response to 6

We have previously shown that the application of double charged negative ion to systems as motivated by the Regge-pole method polarizes the chemical systems such that bond breakage by overcoming binding energies predicted "exactly" accounts for any dispersions, magnetic moments, etc at larger scales and thus accounted for utilizing more fundamental methodology.

#### Response to 7

The application of doubly charged negative ions to the catalysts as predicted initiates complete bond breakage at the (transition states) or activation energies provided in the figures and tables which is what tunes the well-known reaction.

#### Response to 8

The activation barriers are electronic values or electro-volts (eVs) as indicated in all figures and tables.

#### Response to 9

The introduction of double charged negative ions into the chemical systems completely dissociates the reactants as realized by the calculated activation energies as evidenced by "bond breakage" in the transition state.

#### Response to 10

The complete disassociation and bond breakage when double charge negative ion is applied to the initial states highlights the novelty of our approach with potential subsequent elementary step avoidance. We predict that the introduction of double negative ion to the chemical systems tunes the reactions as indicated by DFT.

#### Response to 11

The authors can include the above citings: Our contribution to the above cited works highlight that the key to the fundamental mechanisms underlying all 3 works is indeed due to the addition of 2e-s at initial states of reaction. This can be done via chemical functionalization and substrate modification means as well, however; the introduction of double charged negative ions tends to speed the reactions as well as allow tunability via change of type of catalyst

#### Minor Remark Response

Transition state 1 and Transition 2 labeled as "TS-1" and "TS-2" are the actual activation energy barriers. Initial state 1 and initial state 2, final state 1 and final state 2 are the respective states for transitions labeled "1" and "2"

For further clarification of our well cited fundamental approach please see article:

Single-doped charged gold cluster with highly selective catalytic activity for the reduction of SO<sub>2</sub> by CO: First- Principles Study

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