Computational Investigation of a Reaction Potential Energy Surface

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Background

Much atmospheric and interstellar chemistry involves the reactions of gaseous ions and neutral molecules. Experimental studies of such reactions often include using mass spectrometry to measure the concentration of reactant and product ions. While reactant ion structures can be easily postulated as a result of the methods of preparation, product ion structures (and neutral products) are much more difficult to identify. In most cases, computational chemistry software is used to investigate molecular geometries and energetics of the compounds involved in these reactions. In this study specifically, experimental data obtained at the University of Colorado using mass spectrometric detection is investigated.

Project

In a recent publications[1], it was found that compounds similar to amino acids which contained sulfur were also reactive with excited oxygen. $HSCH_2OO^-$ formed two ionic species and $HSCH_2CO_2^-$ formed three ionic species; both reaction pathways produced neutral products not detected by the mass spectrometer.

The group’s goal was to determine what possible neutral products were produced along with the ionic species observed by mass spectrometry. To do this, the members of the group each chose an ionic product, determined potential non-ionic species with the remaining atoms, and examined the total change in energy of the reaction.

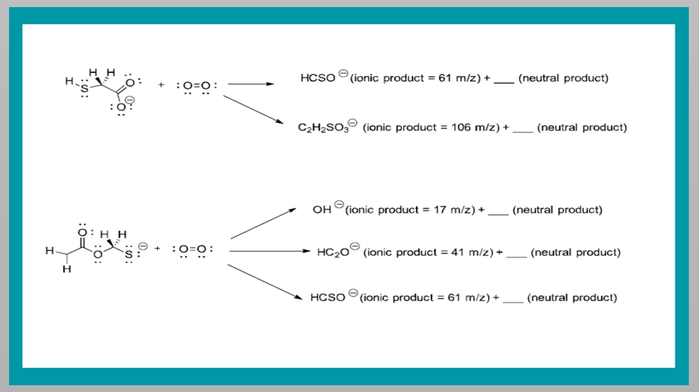
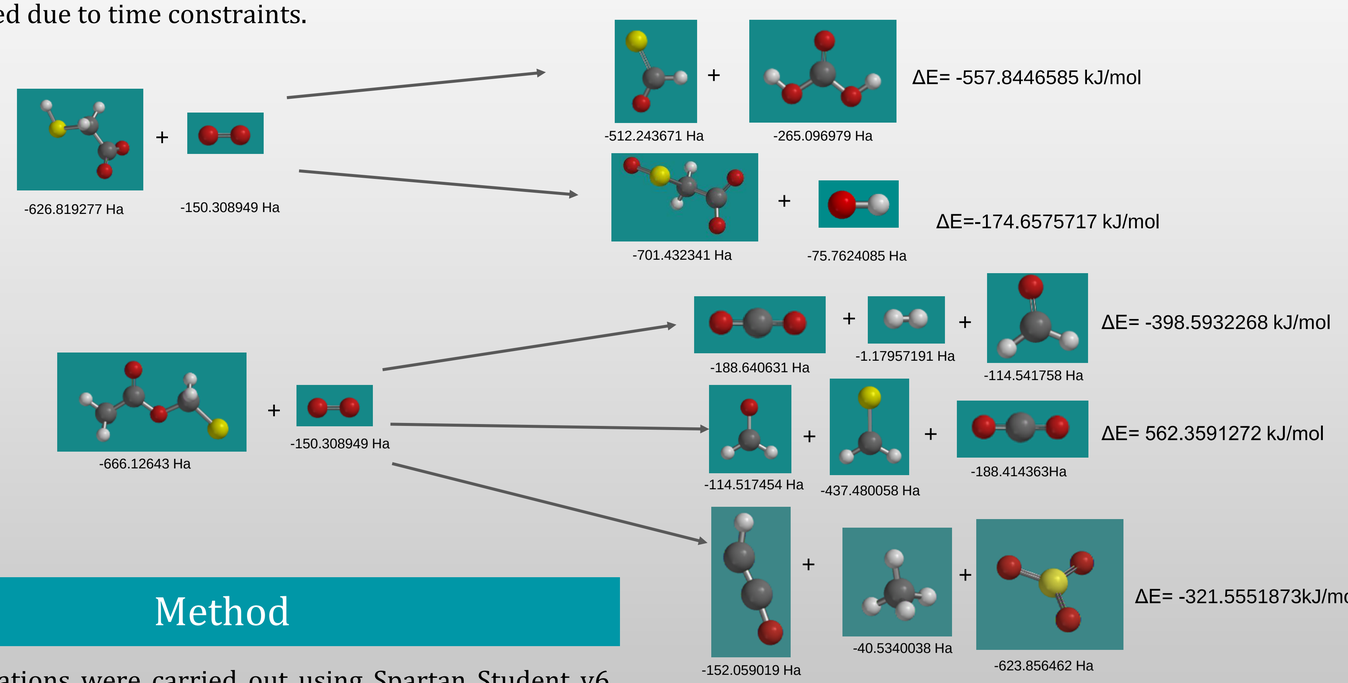


Figure 1: Starting material and ionic products with unknown neutral products

Results

Energy calculations presented here were perfomred using Spartan software. Although Gaussian software was utilized on a supercomputer in an attempt to determine a full reaction pathway that included transition states and enthalpies, the process was not completed due to time constraints.



Method

Energy calculations were carried out using Spartan Student v6. Due to hardware limitations on campus, Gaussian 16 software was run through the San Diego Supercomputer Center Dell Cluster with Intel Haswell Processors (Comet). High performance computing allows for computations that would take days on campus to be completed in minutes with the super computer.

Using the B3LYP level of theory and a 6-311+G(d,p) basis set, calculations for frequency and geometric optimization were made. These methods have been shown to be a good compromise between computational cost and accuracy.[2]

Conclusion

Gas phase ionization chemistry calculations performed the best on the supercomputer rather than the Spartan software. Even though products were predicted using this software it is not definitive that those are the products that are made as there are many potential reaction pathways. Further analysis needs to be done to confirm the products.

References

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