SO$_3$ FORMATION FROM THE IRRADIATION OF SO$_2$ ICE BY X-RAYS: EXPERIMENTS AND INVESTIGATIONS IN THEORETICAL CHEMISTRY

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1. Introduction

Sulfur dioxide (SO$_2$) and its related ions, radicals and fragmentation products are important reagents to prebiotic synthesis, since they can lead to the production of sulfur-containing amino acids. SO$_2$ is also an important constituent of Jupiter moon Io and is also present at other astrophysical environments. This work presents an experimental and theoretical research on the formation of SO$_3$ molecule during the photolysis of a pure SO$_2$ ice sample by synchrotron radiation (mainly soft X-rays) at 12 K.

2. Experimental / Theory

Experiments have been performed at the National Synchrotron Light Laboratory (LNLS), in Campinas, Brazil, under ultra-high vacuum conditions (Pressure below 3 x 10^{-8} mbar). In situ analysis was performed using an infrared Fourier transform spectrometer (FTIR) at different fluences, just as described in details in the experimental work of Pilling and Bergantini [1]. The experimental results led to studies in Theoretical Chemistry. Calculations were performed at MP2 theory level with cc-pVTZ basis set, which is consistent with the studied systems size [2]. All chemical species had the equilibrium geometry confirmed and the enthalpy value obtained at 12 K by applying standard statistical thermodynamics equations. Computations were carried out initially in the gaseous phase and then the solid (SO$_2$ ice). For calculations in the solid phase, the Polarized Continuum Model (PCM) has been chosen [3].

3. Results and Discussions

FTIR spectra of the irradiated SO$_2$ sample have presented formation of SO$_3$, a new chemical species. Through the calculation of reaction enthalpies with or without the solvent influence, one may infer that the presence of the solid SO$_2$, as described by its dielectric constant, decreases the reactivity of the O$^+$ species regarding the formation of SO$_3$. However, it should be noted that SO, O$^+$, O$_2$, and O are products of prior SO$_2$ dissociation (Fig. 1) and more information about the SO$_2$ molecule fragmentation pattern at the experimental conditions would be desirable. Consequently, activation barriers calculations for the most important reactions are in progress. The theoretically identified reaction channels in this study may provide a better understanding for the chemical evolution of SO$_2$-rich ices, which is relevant to both Astrochemistry and Astrobiology.
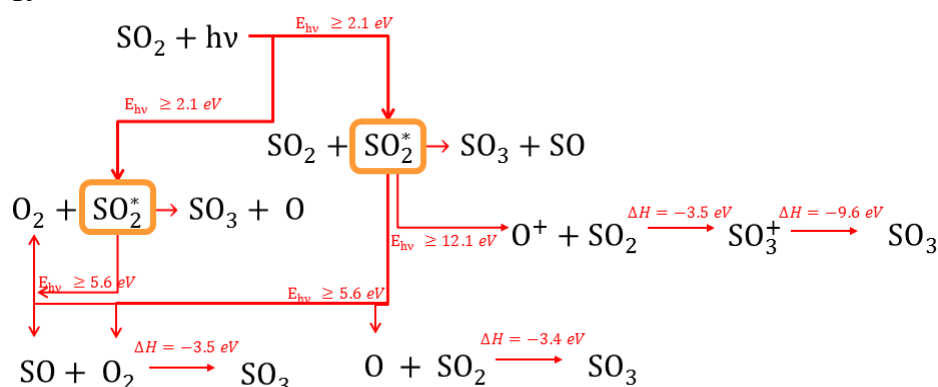
Fig. 1. *Diagram for thermochemically favored SO$_3$ formation routes (in SO$_2$ ice), as indicated from this work calculations. The asterisks in SO$_2$* indicate the chemical species participates as an excited state at the referred chemical reaction, as emphasized by the orange squares.*

4. References

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