1. INTRODUCTION

Hydroxyl radical (OH) is a crucial intermediate in combustion, atmospheric and interstellar chemistry. Although, it is known that these radicals are ubiquitous in the Interstellar Medium (ISM), kinetic data for gas-phase OH-reactions with IS species are still scarce at ultra-low temperatures (10-100 K). Particularly for nitrogen-bearing species, which could form amino acids in the ISM, the literature is very limited.

In this work, the kinetics of the reaction between OH and NH₃ have been studied experimentally at ultra-low temperatures (11.7-177.5 K).

2. EXPERIMENTAL SET-UP AND CRESU TECHNIQUE

The CRESU (French acronym for Cinétique de Réaction en Ecoulement Supersonique Uniforme or Reaction Kinetics in a Uniform Supersonic Flow) technique was used to achieve ultra-low temperatures. It is based on an isentropic expansion of a gas mixture from a relative high-pressure reservoir to a low-pressure chamber through a perfectly designed Laval nozzle (the heart of this technique) which creates a supersonic and cold gas jet uniform in temperature and gas density. This gas mixture is pulsed by a rotary disk which is placed in the divergent part of the Laval nozzle. The pulsed CRESU apparatus developed in our group has been described in detail elsewhere [3].

Pulsed laser photolysis/laser induced fluorescence (PLP-LIF) technique was used to generate OH radicals and to monitor their temporal profile. In this work, the PLP of gaseous H₂O₂ at 248 nm was the source of OH radicals. The LIF from excited OH radicals was detected at ca. 309 nm by a photomultiplier tube as a function of reaction time.

3. KINETIC ANALYSIS

Under pseudo-first order conditions ([NH₃][H₂O₂][OH]₀), the LIF signal from OH radicals decay due to the following reactions:

OH + NH₃ + H₂O₂ → OH + NH₂ + H₂O₂

The pressure dependence of k(T) has been studied at some temperatures. No pressure dependence of k(T) was observed in the density range investigated.

4. EXPERIMENTAL RESULTS OF k(T=11.7-177.5 K)

The results of k(T) determined in the present study are the first experimental determination of k(T) for this reaction in the T<22 K and T=23-177.5 K ranges. These results are shown in a k(T) vs T plot along with previous works (T=230-500 K).

We observe a fast increase of k(T) between 177.5 K and 29.2 K, while k(T) is almost constant in the range 11.7-29.2 K.

However, theoretical calculations predict a decrease in the OH-rate coefficient with temperature. As shown in Figure 4, there is a huge difference between the experimental k(T) and the calculated one by Nguyen and Staton [9].

6. CONCLUSIONS

- Although, there is a huge increase of k(T) in the range 29.2-177.5 K, in general, is continuously increasing when temperature decreases in the explored temperature range (11.7-177.5 K), reaching a maximum around 22 K [4].
- No pressure dependence of k(T) has been observed at the selected temperatures reachable with our current CRESU apparatus.

REFERENCES