## COSMIC RAY EFFECTS IN DIFFERENT ALLOTROPIC WATER ICES

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The ice-mantles is the interface between the carbonaceous and silicates materials, that are very refractory and responsible for building the first grains. This grains possess a rich chemistry that enriches the gas phase.

The surfaces act as a catalyzer of reactions. Condensed molecules at low temperatures react on ices when subjected to radiation field and cosmic rays, forming more complex molecules.



Burke and Brown 2010

Ices are a major reservoir of the heavy elements in several astrophysical environments, extending from cold and dense molecular clouds and protoplanetary disks to the icy bodies in our Solar System, such as comets and Kuiper Belt Objects (Andersson and van Dishoeck, 2008).

Water ice is the dominant constituent of interstellar ices in most lines of sight, being a key molecule in astrochemical models, since it is between the grain and gas phase.

Its many different solid structures depend on the adsorption rate and temperature, and may be amorphous (compact or porous) and/or polycrystalline (cubic or hexagonal).

Amorphous ice at 15 < T < 100 K exists in two forms, a high-density form (Iah) and a low-density form (Ial).

(i) I<sub>ah</sub> ice exists until 38 K;

(ii) between 38 and 80 K,  $I_{ah}$  ice is gradually transformed into  $I_{al}$  ice;

(iii)  $I_{al}$  ice remaining in this form until 100 K;

(iv) Above 100 K, I<sub>al</sub> ice begins to transform into cubic crystalline ice.

Emission of three series of cluster ions,  $(H_2O)_nH^+$ ,  $(H_2O)_nOH^+$ , and  $(H_2O)_n^+$  was present in spectra from water ice at 100 K bombarded by high flux of 15 keV heavy projectiles (collisions in the nuclear regime).

The mass spectra of positive secondary ions ejected from water ice bombarded by a low flux of MeV projectiles (in the electronic regime) are dominated by the series of cluster ions  $(H_2O)_nH^+$ .

With MeV projectiles, the series  $(H_2O)_n^+$ , and  $(H_2O)_nOH^+$  are practically nonexistent; These clusters can be understood as reaction products of the hydronium  $(H_3O)^+$ , and the aggregates  $(H_2O)_n$ ;

So... we prefer to use the notation  $(H_2O)_nH_3O^+$ .

It was showed by Baragiola et al. (using KeV projectiles) and Brown et al. (using MeV projectiles) that sputtering yields are independent of ice temperature up to 60 and 100 K, respectively, but increase at higher temperatures.

Water amorphous ices are commonly found on interstellar grains and continuously bombarded by cosmic rays.

So, the production rate of  $H_3O^+$  and  $(H_2O)_nH_3O^+$  is important for chemistry in interstellar clouds and other environments.

Recent experiments from water ice, performed between 78 and 200 K, revealed strong variations of the secondary ion yield as a function of temperature, which were supposed to originate from phase transitions (Silveira et al., 1999). The same was observed by de Barros et al (2011) using 1.5 MeV  $N_2^+$  beam, between 10 and 216 K.

#### **OBJECTIVES**

In this work, we investigate how the structural changes of water ice influence the emission of secondary ions.

The ion desorption from ices water at different temperatures (30, 70, 100 and 125 and 140 K), was investigated to different allotropic water ices bombarded with heavy ions constituted by Cf fission fragments.

In this way, we study the influence of phase transitions on the desorption yield of  $H_2O$  ions, particularly, the hydronium  $H_3O^+$  and the cluster ions  $(H_2O)_nH_3O^+$ .

#### EXPERIMENTAL SET UP

The experiments have been performed at Van de Graaff ion accelerator located at PUC-Rio, Rio de Janeiro, Brazil.

Inside the high vacuum chamber (base pressure ~  $1x10^{-8}$  mbar), the water vapor was introduced for condensation on a Cu substrate, coupled to a closed-cycle helium cryostat.



#### EXPERIMENTAL SET UP

To simulate the cosmic rays effects, the sample was bombarded by <sup>252</sup>Cf fission fragments. The <sup>252</sup>Cf radioactive source emits FF with energy of about 65 MeV.

Ba<sup>15+</sup> ions are typical <sup>252</sup>Cf FF with  $E/m \sim 0.47$  MeV/u, (one of the most abundant heavy cosmic-ray ions, Fe<sup>*n*+</sup>, has  $E/m \sim 0.36$  MeV/u).

The ions desorbed from water ice thin layers (~ 0.5  $\mu$ m) have been mass/charge analyzed by a time-of-flight spectrometer.



Typical TOF spectrum of the positive desorbed ions from  $H_2O$  condensed at T = 40 K (high density).

The main observed desorbed ions are H<sup>+</sup>, OH<sup>+</sup>, H<sub>2</sub>O<sup>+</sup>, H<sub>3</sub>O<sup>+</sup> and clusters formed by  $(H_2O)_n$  H3O<sup>+</sup>.



For  $H_2O$  condensed at T = 140 K, temperature close of the sublimation, other species arise trapped in the ice.

Condensed at 30 K, the ice is formed by a high density amorphous structure, which density is  $\sim 1,1$  g/cm<sup>3</sup>.

At 70 K, the ice is yet amorphous, but its density is lower, ~ 0,94 g/cm<sup>3</sup>.

To different temperatures have the same (amorphous) phase and them total ion yields are close.

This finding extends the results of Martinez et al. (2006), which show that although the effects of particle bombardment on solid phase molecules depend on the temperature of the sample, the total ion yields, as well as relative ion yields, are significantly different only when phase transition occurs.



The yield distribution of the high density ice (30 K) is different that those of low density ice.

Therefore, we propose that the yield distribution shape could be used as a fingerprint of the ice density.

To n > 14, the yields are almost the same for all ice structures ( $30 \le T \le 125$  K) showing that for large clusters -which are formed in a region away from the impact -the yield is not very sensitive to the sample temperature.

In this work, we used the results at 30 K to study the ion desorption from water ice inside dense interstellar regions.

Despite iron to proton ratio small, the sputtering promoted by heavy ions and energetic ions are 1-2 orders of magnitude higher than ones promoted by the abundant low energy protons (Pilling et al, 2010).

In addition, Andrade et al (2013) showed that heavy ions induce chemical reactions much more efficiently than protons in their respective highest flux region.

In this way, we calculate the number of clusters delivered to gas phase from the water ice at 30 K bombarded by heavy ions.

Table: first column the number of water molecules in the desorbed cluster species and, in the second, the respective ion yield (number of desorbed ions per impact). Last column, an estimative to the ion yield in the interstellar medium. T = 30 K.

Species	Yield	IDR interstellar
$(\mathrm{H_2O})_{n}\mathrm{H_3O^{+}}$	Íon/impact	Ions/s
n = 0	3,79E-3	6,98E-5
n = 1	4,20E-3	7,73E-5
n = 2	3 <mark>,</mark> 68E-3	6,78E-5
n = 3	3,02E-3	5,56E-5
n = 4	2,26E-3	4,16E-5
n = 5	1,89E-3	3,49E-5
n = 6	1,44E-3	2,66E-5
n = 7	1,30E-3	2,40E-5
n = 8	9,10E-4	1,68E-5
n = 9	9,15E-4	1,68E-5
n = 10	7,45E-4	1,37E-5
n = 11	6,00E-4	1,10E-5
n = 12	5,93E-4	1,09E-5

Table: first column the number of water molecules in the desorbed cluster species and, in the second, the respective ion yield (number of desorbed ions per impact). Last column, an estimative to the ion yield in the interstellar medium.

Species	Yield	IDR interstellar
n = 13	- 4,40E-4	8,11E-6
n = 14	3,86E-4	7,11E-6
n = 15	2,61E-4	4,80E-6
n = 16	1,67E-4	3,08E-6
n = 17	1,84E-4	3,40E-6
n = 18	2,24E-4	4,12E-6
n = 19	1,28E-4	2,36E-6
n = 20	1,97E-4	3,62E-6
n = 21	1,45E-4	2,67E-6
n = 22	1,01E-4	1,86E-6
n = 23	1,23E-4	2,26E-6
n = 24	4,43E-5	8,15E-7
n = 25	2,29E-4	4,21E-6
n = 26	4,92E-5	9,06E-7

# Thank you!

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