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► To cite this version:

Sophie Le Caër. Radiolysis of water confined in aluminosilicate nanotubes: importance of charge separation effects. ICIP 2022 - 3rd International Conference on Ionizing Processes 2022, Jul 2022, Idaho Falls (Virtual event), United States. cea-03651501

HAL Id: cea-03651501

<https://cea.hal.science/cea-03651501>

Submitted on 25 Apr 2022

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Radiolysis of Water Confined in Aluminosilicate Nanotubes: Importance of Charge Separation Effects

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Abstract

Imogolite nanotubes are potentially promising co-photocatalysts.¹ Indeed, these clays are described as having a wall polarization allowing them to separate the photo-generated charge carriers. We used here radiolysis as a tool for generating charges and studying their fate.

Two types of aluminosilicate nanotubes were studied: one is hydrophilic on its external and internal surfaces (IMO-OH), while the other has a hydrophobic internal cavity due to Si-CH₃ bonds (IMO-CH₃), the outer surface remaining hydrophilic. Picosecond pulsed radiolysis experiments have demonstrated that electrons are efficiently transferred outside the nanotubes. Coupled to gas production measurements as a function of the water content, these experiments have evidenced that, for imogolite samples containing very few water molecules on the outer surfaces (about 1% of the total mass), quasi-free electrons are formed. They attach to a water molecule, generating a radical water anion, which ultimately leads to the formation of dihydrogen. When more external water molecules are present, solvated electrons, precursors to dihydrogen, are formed.

The attachment of the quasi-free electron to water is a very efficient process, which accounts for the high production of dihydrogen for low values of relative humidity. When the water content increases, the solvation of electrons is predominant compared to the attachment to water molecules. Solvation of electrons therefore leads to the production of dihydrogen, although to a lower extent than when the precursors are quasi-free electrons.

On the other hand, the holes move towards the inner surface of the tubes. They mainly lead to the formation of dihydrogen and methane in irradiated IMO-CH₃.

Our radiolysis experiments therefore demonstrate the wall-induced spontaneous charge separation in these inorganic nanotubes,³ making them very interesting potential co-photocatalysts.

References

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