

Ammonia production, isotopic exchange and sticking on materials relevant to fusion reactors: tungsten and 316L stainless steel

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For high-power operations in ITER, it is foreseen to inject extrinsic impurities into the edge plasma to dissipate part of the plasma exhaust power through radiation and maintain the power fluxes to the plasma-facing components within tolerable limits. To date, the best compromise between radiative efficiency¹ and hot plasma performance^{2,3} has been achieved with the injection of molecular nitrogen (N) in the edge plasma region. However, injection of nitrogen in a divertor plasma environment next to metallic components leads to the conversion of ~10% of the injected nitrogen into NH₃, as it was observed in ASDEX-Upgrade and JET tokamaks during N-seeded discharge^{4,5}. In ITER, ammonia production will be concurrent with a deuterium/tritium plasma thus radioactive tritiated ammonia is expected. The formation of large quantities of tritiated ammonia has consequences for several aspects of the ITER plant operation in terms of tritium retention, gas reprocessing and duty cycle. It is well known that ammonia is a polar molecule and could stick on (shadowed) metallic in-vessel components as well as on pumping ducts. This represents an issue for the tritium recycling plant operated to retrieve tritium from cryopumps and exhaust gases. It is currently unclear how and where ammonia formation predominantly occurs in fusion devices, which makes it difficult to predict the ammonia formation rate in ITER as well as to determine the areas in the vacuum vessel where ammonia might accumulate.

In this contribution, we present new results that will help to model the tritiated ammonia inventory in ITER. In particular, we address the following questions:

- What is the dominant ammonia surface formation mechanism and how does the formation rate depend on the surface material?
- What is the surface assisted isotope exchange efficiency of ammonia?
- What is the sticking probability of NH₃ molecules on ITER-relevant material (tungsten and 316L stainless steel)?

Our studies are performed in an ultra-high-vacuum environment⁶ using ion beam and molecular beam exposures of polycrystalline tungsten (W) and 316L stainless steel (316L-SS) samples probed by various mass spectrometry and spectroscopic methods.

To understand the mechanism of ND₃ formation from elements implanted in the material bulk, sequential implantation of N₂⁺ and D₂⁺ ions at 300 K is performed and Temperature Programmed Desorption (TPD) is used to quantify HD, D₂, N₂ and ND₃ production rates. On the one hand, we show that deuterated ammonia (ND₃) is produced on both metals when bulk deuterium (D) diffusion is activated. On the other hand, the absolute quantity of ND₃ produced is found to be strongly dependent on the sample material. This difference in ND₃ production rate is related to dissimilarities in the formation process. On polycrystalline tungsten, the formation mechanism involves the implanted N atoms that are present at the surface. Nitrogen implanted deeper in tungsten is unable to participate in the production of ND₃. In stark contrast, on 316L-SS, N atoms naturally contained in the bulk of the alloy participate significantly in the formation of ND₃ through diffusion to the surface, which occurs in the temperature range investigated here (300 –800 K).

The surface assisted production of ammonia isotopologues NH_{3-x}D_x has been studied with two complementary approaches. To quantify the efficiency of isotope exchange between neutral species, a molecular beam of NH₃ molecules impinging on a deuterated tungsten surface has been analyzed by on-line mass spectrometry. To evaluate which edge plasma species is most efficient for surface assisted isotope exchange, NH_x surface precursors have been exposed to either, a neutral D₂ flux, an ionic D₂⁺ flux or an atomic D flux and the resulting surface precursors have been probed by High Resolution Electron Energy Loss Spectroscopy. It is found that D radicals are most efficient while D₂ neutral fluxes are least efficient.

Finally, the absolute sticking probabilities of NH₃ molecules on W and 316L-SS surfaces have been measured with the molecular beam reflectivity method of King & Wells⁷. For each material, three parameters have been varied independently: the material surface temperature, the NH₃ kinetic energy and the NH₃ coverage on the material surface. Firstly, the sticking probability of NH₃ decreases exponentially from near unity down to a few percent when increasing the material surface temperature from 125 to 425 K. Secondly, ammonia sticking probability is divided by a factor of ~3 when increasing the NH₃ kinetic energy from 55 meV up to 255 meV. Thirdly, NH₃ sticking probability exhibits a complex behavior as a function of NH₃ coverage on the surface: the sticking probability initially increases and then decreases. Finally, NH₃ dissociation is found to be negligible below 450 K as seen by in situ Auger Electron Spectroscopy. These experimental observations

reveal a molecular adsorption of ammonia on both W and 316L-SS that is assisted by two surface precursors, a NH₃ molecule bouncing on the bare surface (intrinsic precursor) and a NH₃ molecule bouncing atop a NH₃ covered area of the surface (extrinsic precursor).

A Generalized and Separable Kisliuk (GSK) kinetic model describing this precursors-mediated adsorption has been developed and it is able to reproduce quantitatively the evolution of NH₃ sticking probabilities with NH₃ surface coverage and the material surface temperature. The departure of the experimental data to the GSK model at the highest coverage (Figure 1) is explained by a long living (~ second lifetime) extrinsic precursor which manifests itself by a NH₃ partial pressure spike when the molecular beam exposure is stopped.

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