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Ammonia production on tungsten and stainless steel during nitrogen seeded H (D) plasmas in the linear plasma device GyM

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Divertor tokamaks using all-metal plasma-facing components need impurity seeding of nitrogen (N2), neon (Ne), or argon (Ar) for heat load control under high power conditions. Considering present day devices (i.e. AUG), N2 is compatible with both divertor dissipation and good core performance [1].
However, the formation of tritiated ammonia due to chemical reactions between N2 and fuel isotopes may pose challenges in the tritium inventory of future nuclear fusion power plants like ITER, by increasing in-vessel retention and impacting the gas-handling plant [2]. A detailed quantification of the ammonia produced during tokamak nitrogen-seeded plasmas is therefore crucial.
To address this critical point, nitrogen-seeding experiments were performed in our laboratory by using the fusion-relevant linear plasma device GyM [3].
Since the formation of ammonia in fusion-relevant plasmas is a plasma-catalysis process [4], we investigated the effect of both the surface wall material (different chemical behaviour and roughness (Ra) of the tungsten and stainless steel) and the hydrogen isotope (hydrogen or deuterium) on the ammonia production.
GyM is made up of a cylindrical stainless steel (SS) vacuum vessel (Ra ≤ 0.6 μm), in which an additional tungsten (W) liner (Ra ≈ 10μm) can be inserted. Walls are heated by plasma and temperature is measured by two thermocouples. After about 30 min of plasma, wall temperature reaches a constant value of 400 K. Experiments were performed keeping constant the N2/D2 partial pressure ratio at 10% and varying the nitrogen concentration from 2% to 10%; in this conditions the neutral plasma pressure varies between 2.0x10-2 Pa to 5.5x10 −2 Pa. The main plasma parameters were Te=5eV and ne=2 x 1016 m-3.
Residual gas analyser (RGA) and optical emission spectroscopy (OES) diagnostics were both used during the experiments for qualitative analyses of gas species and radicals resulting from the plasma. In particular, OES detects the emission bands of ND radicals at 335.7 nm and 336.4 nm. Assuming that these signals come from ammonia formation, band intensity can be used as an indicator of how much ammonia is produced during the experiments. In order to quantify the produced ammonia the exhaust from GyM vessel is collected by a liquid nitrogen trap (LN trap). Ammonia contained in LN trap is then counted by a chromatographic system.
Results, in terms of ammonia conversion yield on a W wall, were compared with those obtained in the same experimental conditions, with a SS wall. Chromatographic analysis showed that more ammonia was produced during experiments with W wall (fig.1) and this is also validated by OES (fig.2). Ammonia conversion was 17% on SS and increased until 22% on W.
This behaviour in presence of the tungsten liner was expected. In fact N atom possesses an electron lone pair available for a true chemical covalent bond, and the strength of this bond is strictly linked to the empty atomic orbitals of the surface. Since W is a higher electron acceptor than the elements of SS, the reaction rate associated to the ammonia formation is increased because N reacts faster with W. Catalytic phenomena like ammonia formation on a metal surface are also affected by the surface roughness. A rise of it produces an increase in the surface area available for chemical reactions (active sites). Therefore the number of the reactions that take place simultaniously increases and the ammonia production increases too.
Pressure-dependent ammonia formation was also found on W and SS.
A series of experiments was performed to evaluate the isotopic effect on the ammonia formation in GyM with the SS wall. Results from chromatography and OES showed that the ammonia formation was higher in hydrogen-seeded plasmas (fig.3).
The isotopic effect observed is explained taking into account that a bond involving a D atom is less reactive than the analogous bond involving an H atom; D containing species are typically more stable and exhibit lower reaction rate constants than comparable hydrogen containing isotopologues.
The results confirm that a catalytic process is actually involved in ammonia production. In particular, the active species (radicals) are formed in the plasma volume and ammonia formation takes place on the surface of the wall that acts as catalyst.
Our work also demonstrates that a system composed by liquid nitrogen trap and chromatograph allows to absolutly quantify the ammonia contained in the exhaust of GyM. This strategy has already been applied to AUG tokamak, too.
Next step in this field is to demonstrate that it is possible to recover hydrogen isotopes from ammonia cracking. To this aim experiments are now ongoing. The exhaust of GyM is again collected by a liquid nitrogen trap. It is then chemically analyzed by gas chromatography. It eventually flows in a chemical reactor in which catalytic cracking of the ammonia occurs.

[1] C. Giroud et al, Nucl. Fusion 52(2012) 063022
[2] R. A. Pitts et al, Nuclear Materials and Energy 20 (2019) 100696
[3] L. Laguardia et al, Nuclear Materials and Energy 12 (2017) 261266
[4] Marwa Ben Yaala et al, Phys. Chem. Chem. Phys., 2019, 21, 16623

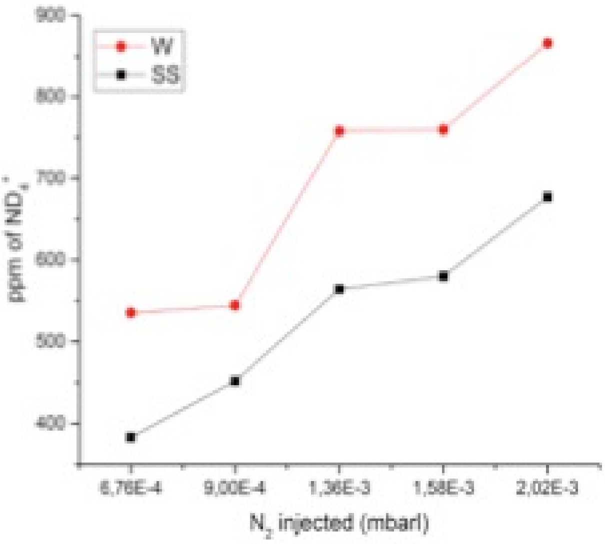

Figure 1: Ammonia content in the exhaust collected during each N seeded experiment. Experiments differ from each other for the nitrogen flux injected in D plasmas.

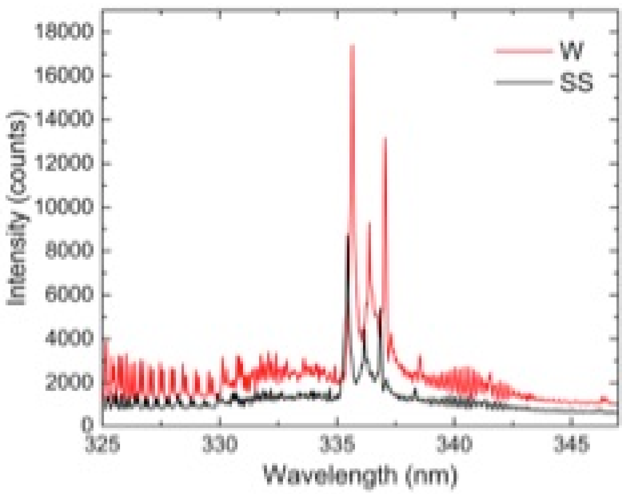

Figure 2: OES spectrum showing (335.7 nm and 336.4 nm) ND and N2 (337.1 nm) bands

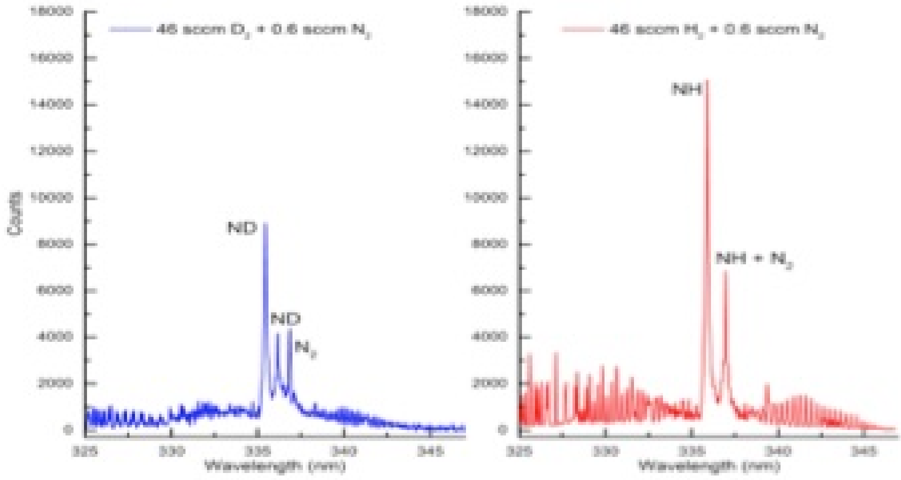

Figure 3: OES spectra acquired during N seeded D plasma (a) and N seeded H plasma (b)

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