Nitrogen balance and ammonia formation during nitrogen seeded discharges at ASDEX Upgrade

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The intrinsic radiation in the scrape off layer plasma of present all-metal fusion devices is too low to prevent overheating of plasma-facing components (PFCs) during high power discharges. In ASDEX Upgrade (AUG) seeding of gaseous impurities is used to protect the tungsten divertor from local heat loads [1]. The best candidate seems to be nitrogen, but chemical reactions with hydrogen, i.e., the formation of ammonia, are observed [2]. For future fusion devices ammonia can be a serious issue for gas handling plants and cryo pumps. Furthermore, in AUG plasmas increased nitrogen concentrations are observed in non-seeded discharges subsequent to discharges with N2 seeding. To investigate the role of the tungsten PFCs with respect to nitrogen storage the residual gases of discharges with and without nitrogen seeding at AUG have been investigated by mass spectrometry.

Evaluation of mass spectra

At AUG quadrupole residual gas analyzers (RGAs) are mounted at the pumping ducts. To enable an absolute evaluation of the spectra they are combined with calibrated capacitance manometers and ionization gauges. For an absolute calibration the sensitivity of the instruments, the individual cracking patterns and the pumping speed have to be determined. To overcome this problem a calibrated test gas mixture with known composition was used was produced. It contains methane (9,79%), ammonia (9,88 %), ethane (10,59 %), argon (10,09 %), D (15,38 %) and helium (44,37 %). In the pumping ducts the pressure is in the transition range, i.e. there is a non-negligible interaction between the main gas deuterium (D) and the impurity gases [3]. Therefore, a significant amount of light gases was added to the test gas. It is a challenge to distinguish the deuterated species of water, methane and ammonia by mass spectrometry as they all produce a dominant peak at a mass-to-charge ratio of 20 amu/e. Therefore, accurate knowledge of the cracking pattern is needed for decomposition of the spectra. These patterns were derived using a statistical approach from the measured un-deuterated data [3].

Residual gas composition

Exemplary results from the residual gas analysis after decomposition of the spectra for similar discharges with and without N_2 seeding are shown in Fig.1. Absolutely calibrated data during and after the plasma discharges are given for methane, water, nitrogen and ammonia. For the non-seeded discharge the total pressure during the discharge is 0.1 Pa and the typical fractions of impurities in the pumped gas are in the low percent range. During the discharge almost steady state conditions are reached. After the plasma discharge a fast decay is observed for all species. In the seeded case, a continuous rise of the N_2 content in the residual gas is found during the discharge, even though the seeding rate is constant. In contrast, only a small amount of ammonia is observed in this phase while strong ND_3 outgassing is found just after the discharge, when ammonia dominates the impurities.

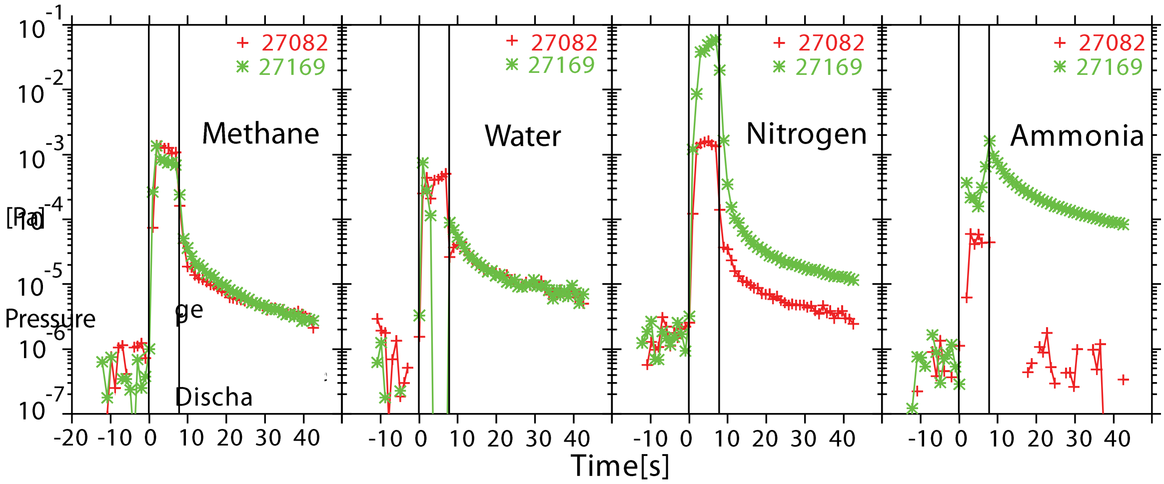

Figure 1: Partial pressures for a non-nitrogen seeded (27082) and a seeded (27169) H-mode plasma discharge.

Nitrogen balance

Absolute calibration of the RGA and the knowledge of the pumping speeds [4] allow to determine the absolute amounts of pumped gas. For the first discharge of a series of three identical N_2-seeded H-mode discharges, 33 % of the nitrogen injected is missing (Fig. 2). For the second discharge this fraction decreases to 19 % and for the third one the balance is almost closed. This can be understood assuming that the missing nitrogen is stored at the PFCs and that this stored inventory saturates within 3 discharges. The ammonia behaves quite opposite: whereas for the first discharge only 2 % of the seeded nitrogen is observed as ammonia, this value rises to 5 % at the last discharge of this series. The role of ammonia on the storage of nitrogen at the PFCs is confirmed by a second set of discharges: A series of comparable discharges this time not only with but also without nitrogen seeding was performed. The measured amounts of N_2 and ND_3 resulting from the

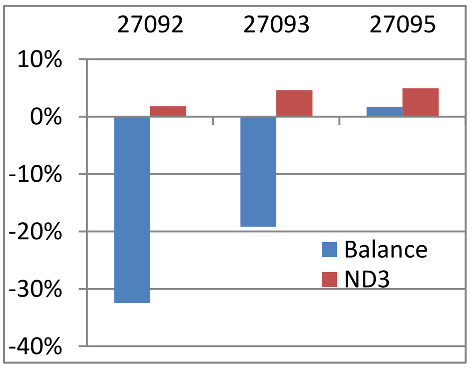

Figure 2: Nitrogen balance and ammonia production for 3 identical subsequent plasma discharges

RGA decomposition are shown in Fig.3. Nitrogen was seeded only during the discharges 27169 and 27172 of this series. Large amounts of nitrogen are pumped during these seeded discharges. It is reasonable to assume that this fraction is pumped directly, without reaching the PFCs, as the gas was seeded in the divertor region. Again only small amounts of ammonia are observed during the discharges, but the amount of ammonia after the discharge is almost the same for seeded and non-seeded discharges. The first seeded discharge seems to build up a wall inventory of ammonia, which can be released in subsequent plasma discharges.

The rise of the nitrogen signal during a discharge with constant gas puff, the pumping of ammonia after discharges and the strong ammonia outgassing in non-seeded discharges subsequent to discharges with seeding indicate the role of ammonia on the storage of N_2 at the vessel PFCs.

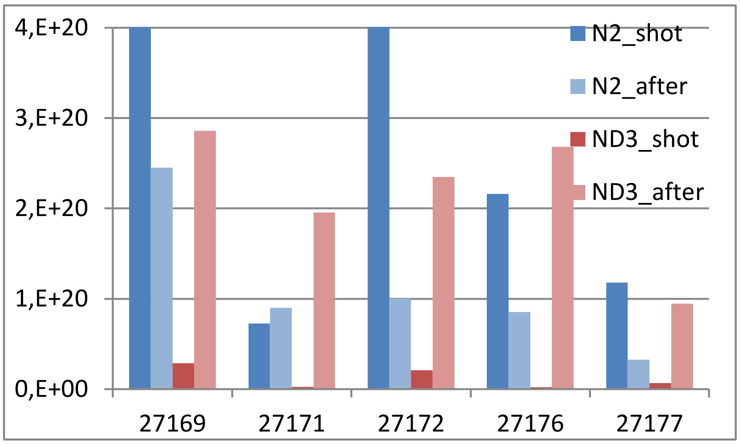

Figure 3: Pumped nitrogen and ammonia integrated during and after the plasma discharge for a series of discharges with (27169,27172) and w/o nitrogen seeding (27171,27176,27177). The maxima are clipped.

Sticking of ammonia to the PFCs and isotope exchange reactions

As mentioned above, for the calibration of the residual gas analyzers a test gas mixture, which contains almost the same amount of methane and ammonia was used. As non deuterated species were used, ammonia and methane could be distinguished by their main peaks at 17 and 16 amu/e. Equal peak intensities for both species should be expected upon injection of the mixture. However, 85 % of the ammonia was missing at the start of the gas inlet. Apparently, a large part of the injected ammonia is pumped by the PFCs in AUG. With time the ammonia signal rises, indicating the saturation of the wall by ammonia. Parallel to the rise of the ammonia peak at 17 amu/e a peak at 18 amu/e appears. As the PFCs at AUG are saturated with D, this signal can be interpreted as the appearance of NH_2D, which is produced at the quadrupole or at the PFCs.

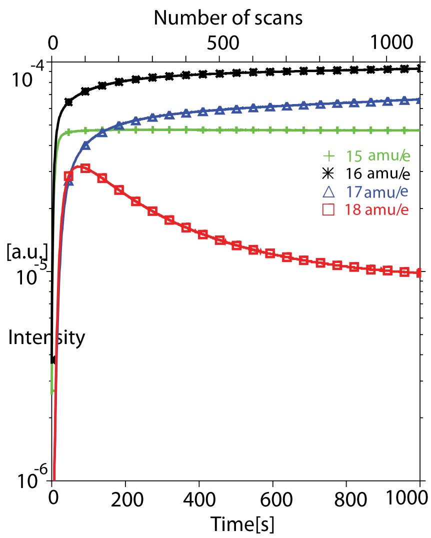

Figure 4: Residual gas in a laboratory device after inlet of the test gas, when the wall was filled up with D.

Since a detailed investigation of this effect was not possible at AUG, a simple laboratory experiment was performed. The identical residual gas analyzer was installed at a stainless steel vessel with a volume of 500 l. The installed pumping speed was adjusted to yield a similar ratio of pumping speed to surface area as at AUG. After injecting the above test gas mixture again the majority of ammonia disappears, but no peak at 18 amu/e appears. After preloading the wall with a D_2 glow discharge the experiment was repeated. The results are shown in Fig. 4. The peak at 15 amu/e is produced by methane only, 16 amu/e by methane and ammonia and 17 amu/e by ammonia only. Upon injection of the test gas the methane signal immediately reaches steady state, whereas the integrals of the peaks at 16 amu/e and 17 amu/e need about 1000 s to stabilize. According to the composition of the test gas and the electron ionization cross sections equal peak intensities at 17 amu/e and 15 amu/e were expected. However, as in AUG, the majority of the ammonia is missing at the beginning of the inlet.

Just as in AUG, with the vessel walls preloaded with deuterium the additional peak at 18 amu/e appears. At first its intensity is equal to the one at 17 amu/e (representative for ammonia), but soon it starts to decrease with the same time constant that describes the further rise of the ammonia peak. This behavior hints to the role of surface processes at the metal walls: As proposed in [5] ammonia seems to stick to the surface as NH_2 after having lost one hydrogen atom. It is then released after a reaction with the surface bound deuterium, resulting in the production of NH_2D, which accounts for the signal at 18 amu/e.

Interpretation

From the results presented above we try to derive a qualitative picture of the processes involved (Fig. 5). As shown in Fig 2, most of the nitrogen seeded is pumped as N_2 during the plasma discharge. Ionized nitrogen in a deuterium environment will form finally ammonia. From the test gas injection we assume that, without plasma, 85 % of this ammonia is pumped by the PFCs. As the ammonia is easily ionized and strongly pumped by the wall only small amounts of ammonia are observed during plasma discharges. The majority of ammonia is stored at the surface. The production of NH_2D, hint to the role of chemical processes [5]. In between discharges part of the ammonia is released and pumped. However, parts of the PFCs remain saturated with ammonia. For this reason the fraction ammonia storage is reduced in the next seeded discharge and the nitrogen retention decreases from discharge to discharge. This also explains the fact that the ammonia production is similar for seeded and non-seeded discharges. The storage as ammonia also explains the medium term retention of nitrogen, which hinders the control of the nitrogen radiation in subsequent plasma discharges [1].

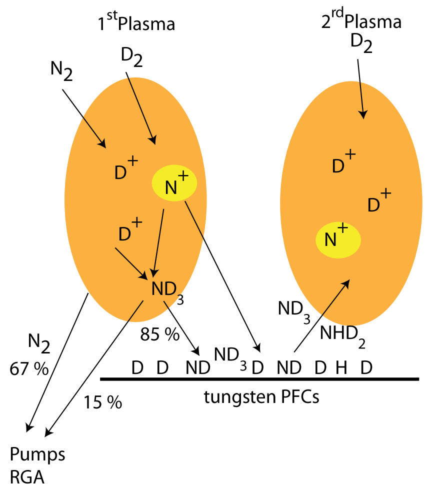

Figure 5: Sketch of the reactions for a nitrogen seeded plasma discharge.

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