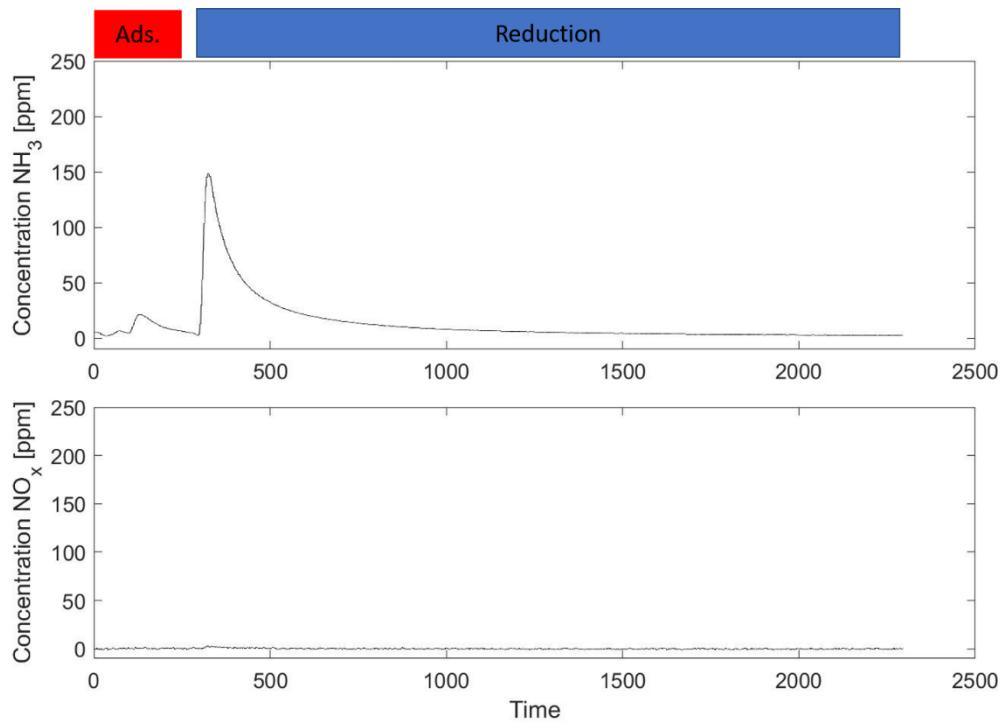


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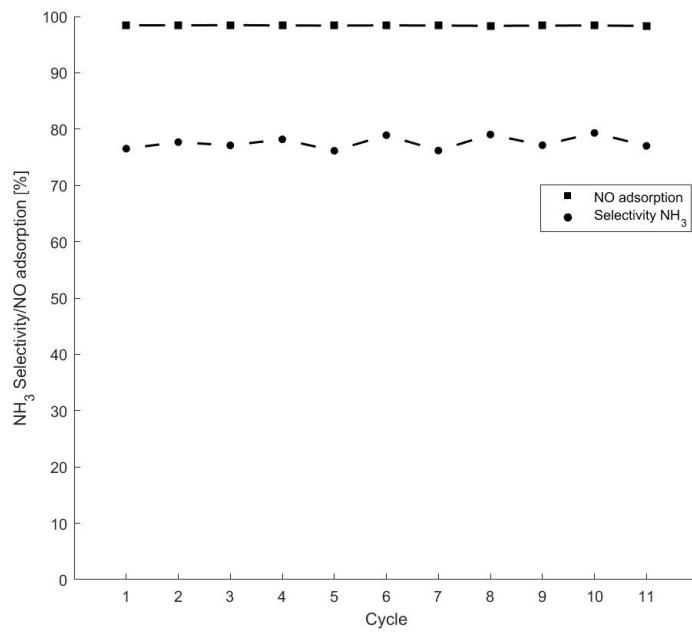
## Supporting Information

### **Energy-Efficient Small-Scale Ammonia Synthesis Process with Plasma-enabled Nitrogen Oxidation and Catalytic Reduction of Adsorbed NO<sub>x</sub>**

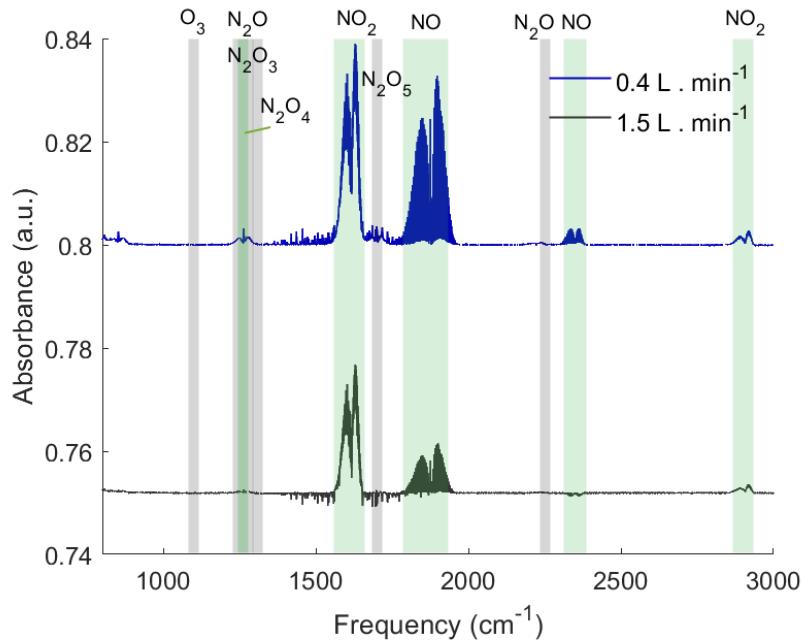
Lander Hollevoet, Elise Vervloessem, Yury Gorbanev, Anton Nikiforov, Nathalie De Geyter, Annemie Bogaerts,\* and Johan A. Martens\*



**Figure S1.** Outlet concentration of  $\text{NH}_3$  and  $\text{NO}_x$  during an adsorption-reduction cycle at 175 °C. Adsorption phase was executed with a gas mixture composed of 200 ppm NO, 20 %  $\text{O}_2$  and 80 %  $\text{N}_2$ , reduction phase with a gas mixture composed of 1000 ppm  $\text{H}_2$  in  $\text{N}_2$ .  $\text{NH}_3$  selectivity = 68 %



**Figure S2.** Stability of the LNT catalyst during 11 consecutive adsorption/reduction cycles at 175°C. Adsorption phase was executed with a gas mixture composed of 200 ppm NO, 20 %  $\text{O}_2$  and 80 %  $\text{N}_2$ , reduction phase with a gas mixture composed of 5%  $\text{H}_2$  and 95 %  $\text{N}_2$ .



**Figure S3.** The FTIR spectra after the plasma for the highest concentration (blue; 0.4 L/min) and the lowest energy consumption (black; 1.5 L/min). The peak regions of the detected species are annotated in green ( $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{N}_2\text{O}_4$ ), and the absent species in grey ( $\text{O}_3$ ,  $\text{N}_2\text{O}$ ,  $\text{N}_2\text{O}_3$ ,  $\text{N}_2\text{O}_5$ ).

**Table S1.** Overview of the best performing small-scale processes for decentralized NH<sub>3</sub> production

Small-scale NH <sub>3</sub> synthesis concepts					
	Yield	NH <sub>3</sub> selectivity	Energy cost	Max. NH <sub>3</sub> conc.	Ref.
Plasma NO <sub>x</sub> + catalytic reduction	Plasma: 1 mmol/h LNT: 10.2 mmol/h.g <sub>Pt</sub> 0.077 mmol/h.g <sub>LNT</sub>	Selectivity = 83 %	2.1 MJ/mol NH <sub>3</sub> *	240 ppm	This work
Electrochemical N <sub>2</sub> reduction with Li-cycling	0.19 mmol/cm <sup>2</sup> .h	FE = 69 %	2.9 MJ/mol NH <sub>3</sub> *	/	1
Electrochemical N <sub>2</sub> reduction with Li-cycling	0.11 mmol/cm <sup>2</sup> .h	FE = 35 %	12.4 MJ/mol NH <sub>3</sub>	77 ppm	2
Plasma NO <sub>3</sub> <sup>-</sup> + Electrochemical reduction	Plasma: 0.8 mmol/h Elec.: 0.15 mmol/h.cm <sup>2</sup>	FE =~100 %	15.5 MJ/mol NH <sub>3</sub>	60 ppm	3
Plasma NO <sub>3</sub> <sup>-</sup> + Electrochemical reduction	Plasma: 1.35 mmol/h Elec.: 0.11 mmol/h.cm <sup>2</sup>	FE = 95.8 % Selectivity = 81.2 %	/	75 ppm	4
Direct Plasma NH <sub>3</sub>	4 mmol/h	Selectivity = ~ 100 %	19.1 MJ/mol NH <sub>3</sub> *	1.75 %	5

\*Assuming H<sub>2</sub> is produced at an energy efficiency of 70 %

**Table S2.** Comparison of our Pt/BaO/Al<sub>2</sub>O<sub>3</sub>Lean NO<sub>x</sub> Trap (heterogeneous catalyst) with state of the art electrocatalysts for reduction of nitrate ions to NH<sub>3</sub>

Conversion of oxidized nitrogen (NO <sub>x</sub> /NO <sub>3</sub> <sup>-</sup> ) to NH <sub>3</sub>				
	Catalyst	NH <sub>3</sub> yield	NH <sub>3</sub> selectivity	Ref.
Catalytic NO <sub>x</sub> reduction	Pt/BaO/Al <sub>2</sub> O <sub>3</sub> Lean NO <sub>x</sub> Trap	0.077 mmol/h.g <sub>LNT</sub> 10.2 mmol/h.g <sub>Pt</sub>	Selectivity = 83 %	This work
Electrochemical nitrate reduction	Cu nanowires	0.15 mmol/h.cm <sup>2</sup>	FE = ~ 100 %	3
	Pd/TiO <sub>2</sub>	0.16 mmol/h.cm <sup>2</sup>	FE = 92.1 %	6
	Single atom Fe	0.075 mmol/h.cm <sup>2</sup>	FE = ~ 100 %	7
	Single atom Fe	0.44 mmol/h.cm <sup>2</sup>	FE = 75 %	8
	Ni/B	0.11 mmol/h.cm <sup>2</sup>	FE = 98.7 %	4
	Cu/Cu <sub>2</sub> O nanowires	0.24 mmol/h.cm <sup>2</sup>	FE = 95.8 % Selectivity = 81.2 %	9

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