**Supplementary Material**

**Competitive Interplay of Deposition and Etching Processes**

**in Atomic Layer Growth of Cobalt and Nickel Metal Films**

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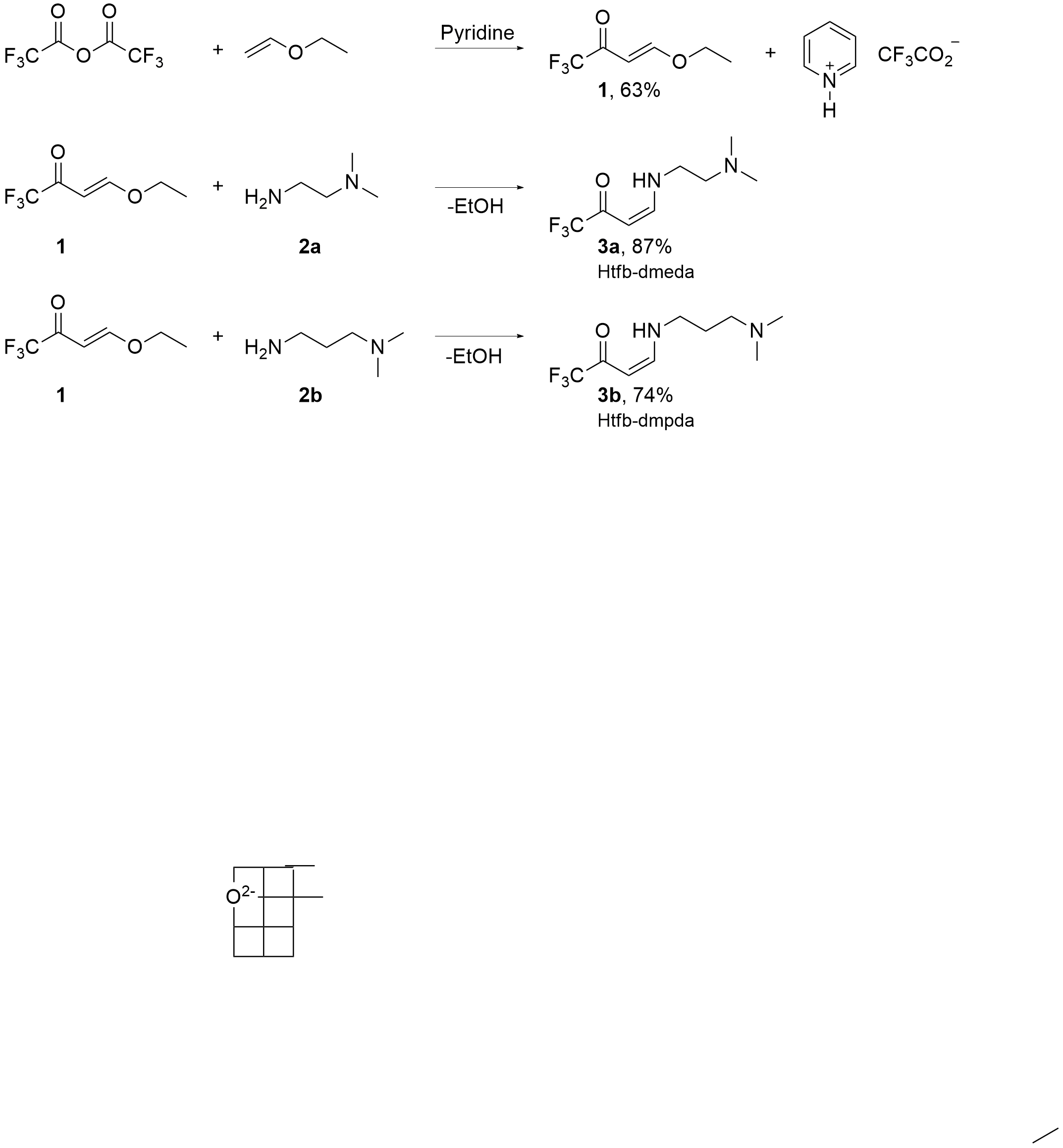
**S6** XRD patterns of Co and Ni films after annealing under argon……………………. S8

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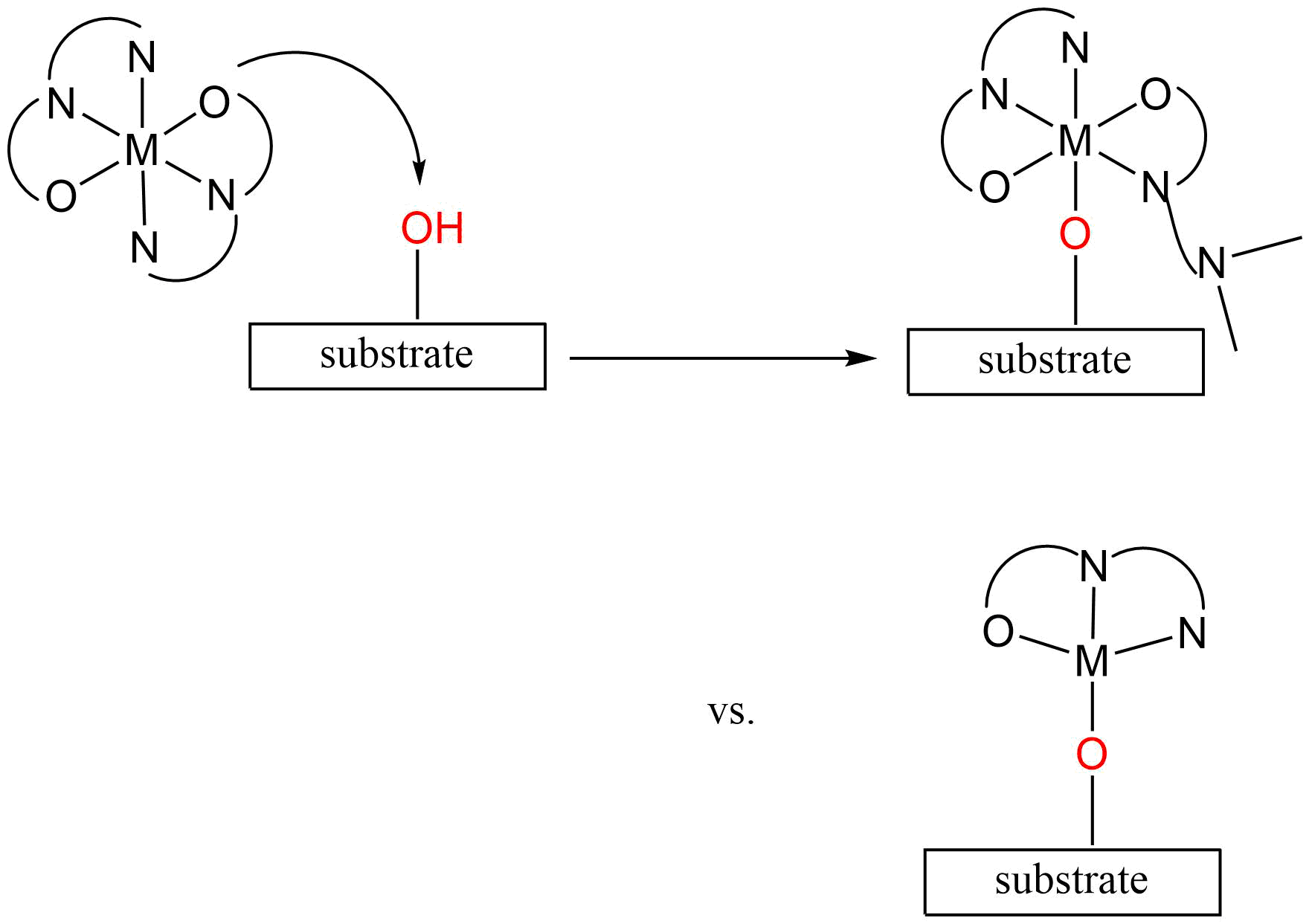
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**Table SI:** F-F distances and sublimation temperatures of precursors **4a**, **4b**, **5a** and **5b**.

|  |  |  |
| --- | --- | --- |
| Compound | F-F distance  [Å] | Sublimation temperature [°C] / 10-3 mbar |
| **4a** | 3.06 | 170 |
| **5a** | 3.04 | 180 |
| **4b** | 3.12 | 140 |
| **5b** | 3.08 | 150 |

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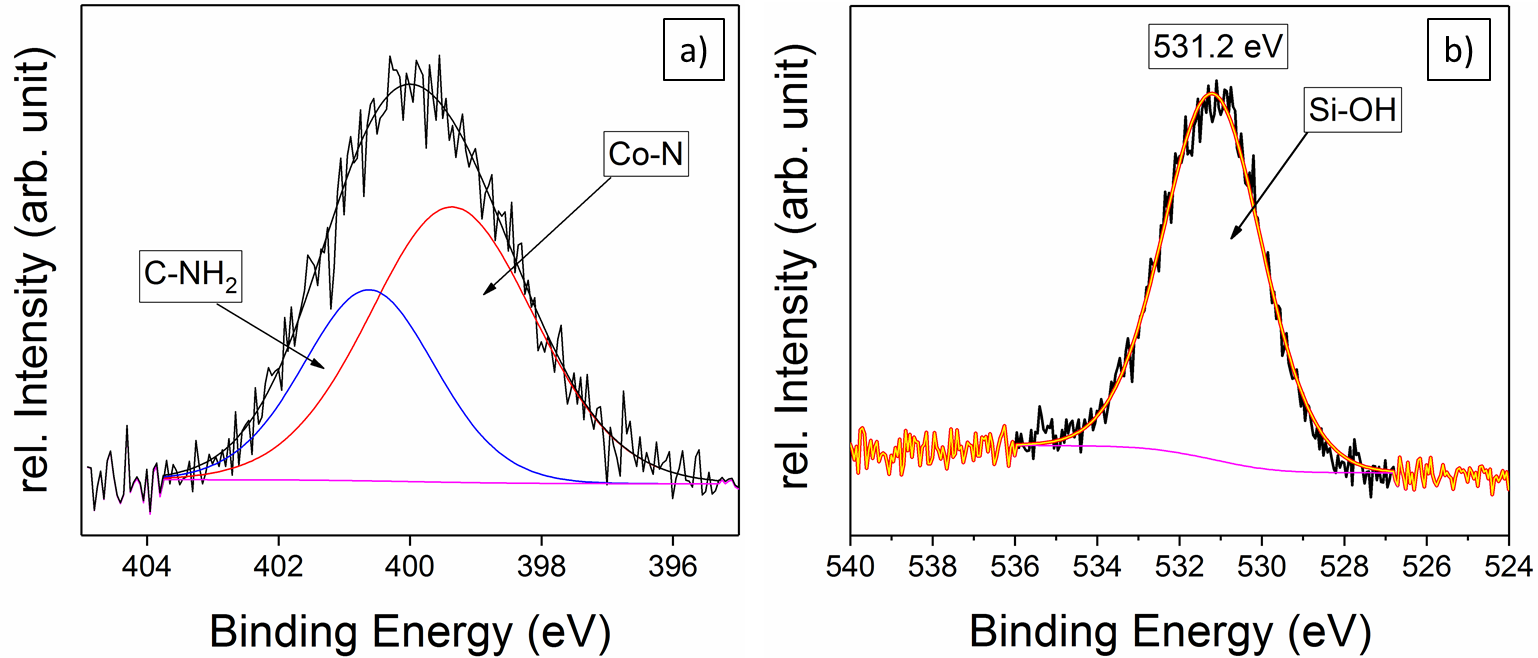
**Scheme S1:** Two-step synthesis of tridentate enaminone ligands **3a** and **3b**.



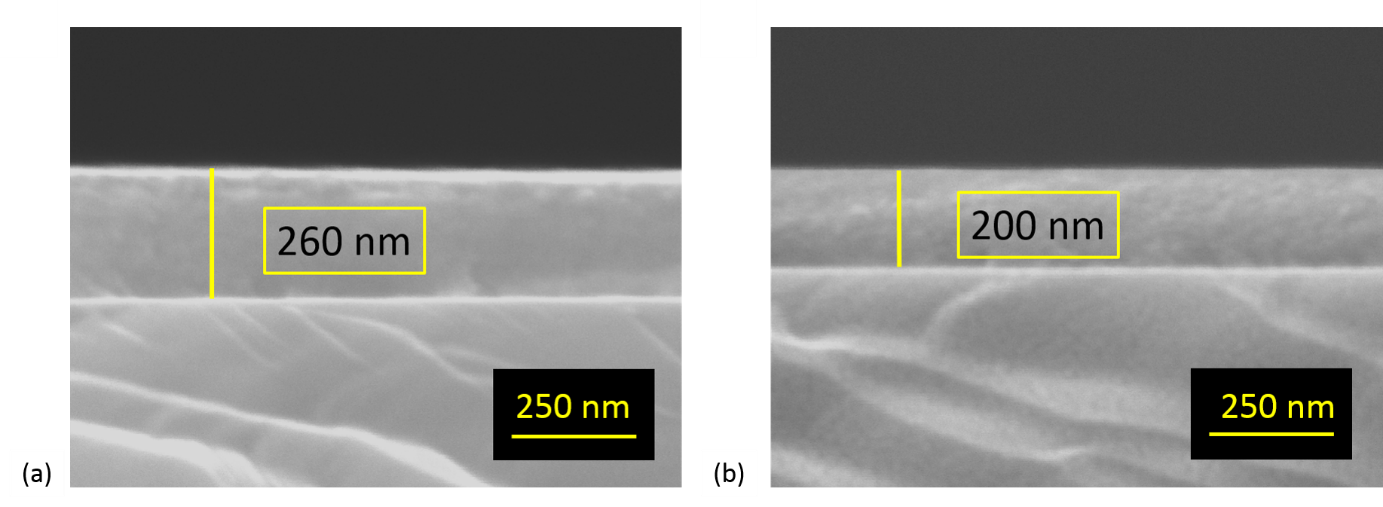
**Scheme S2:** Schematic drawing of the precursor binding mode upon chemisorption onto the substrate surface.



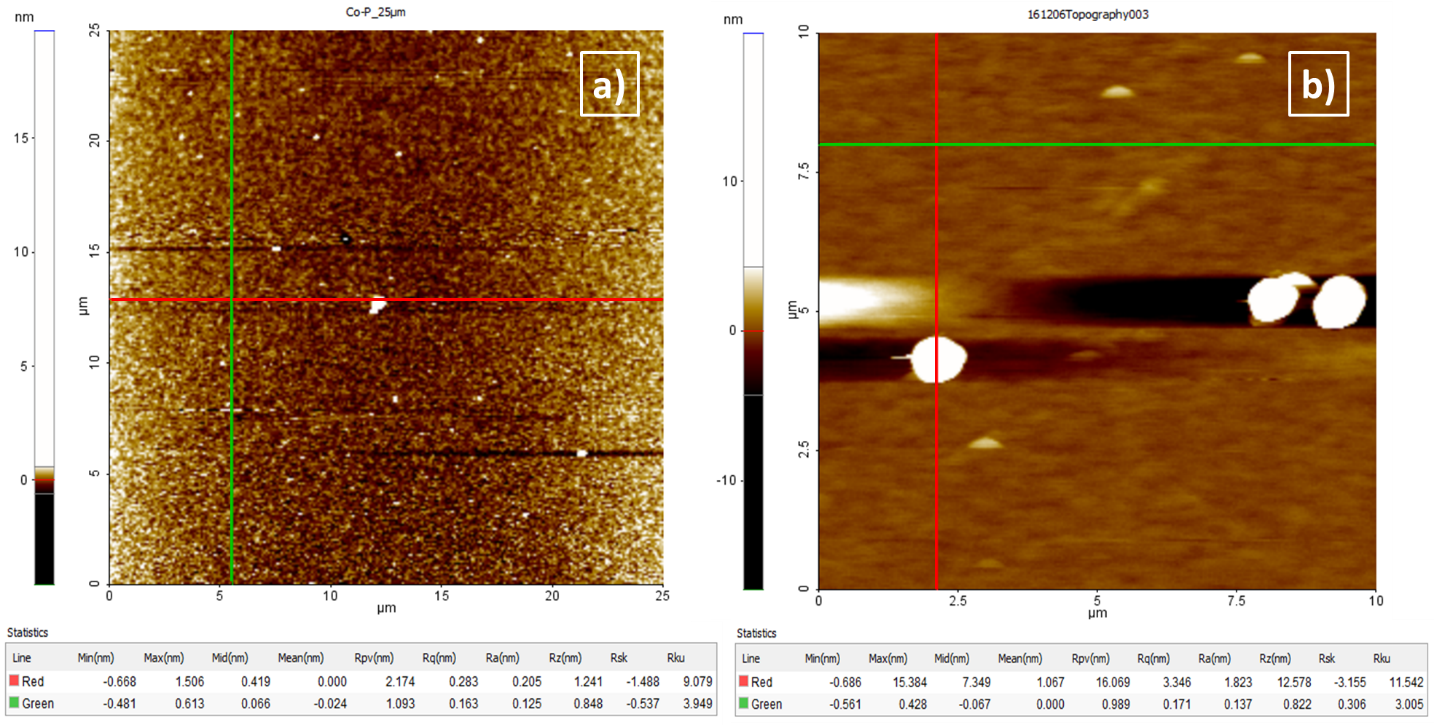
**Scheme S3:** Schematic illustration of the role of hydrogen plasma as activant and etchant during the ALD process.



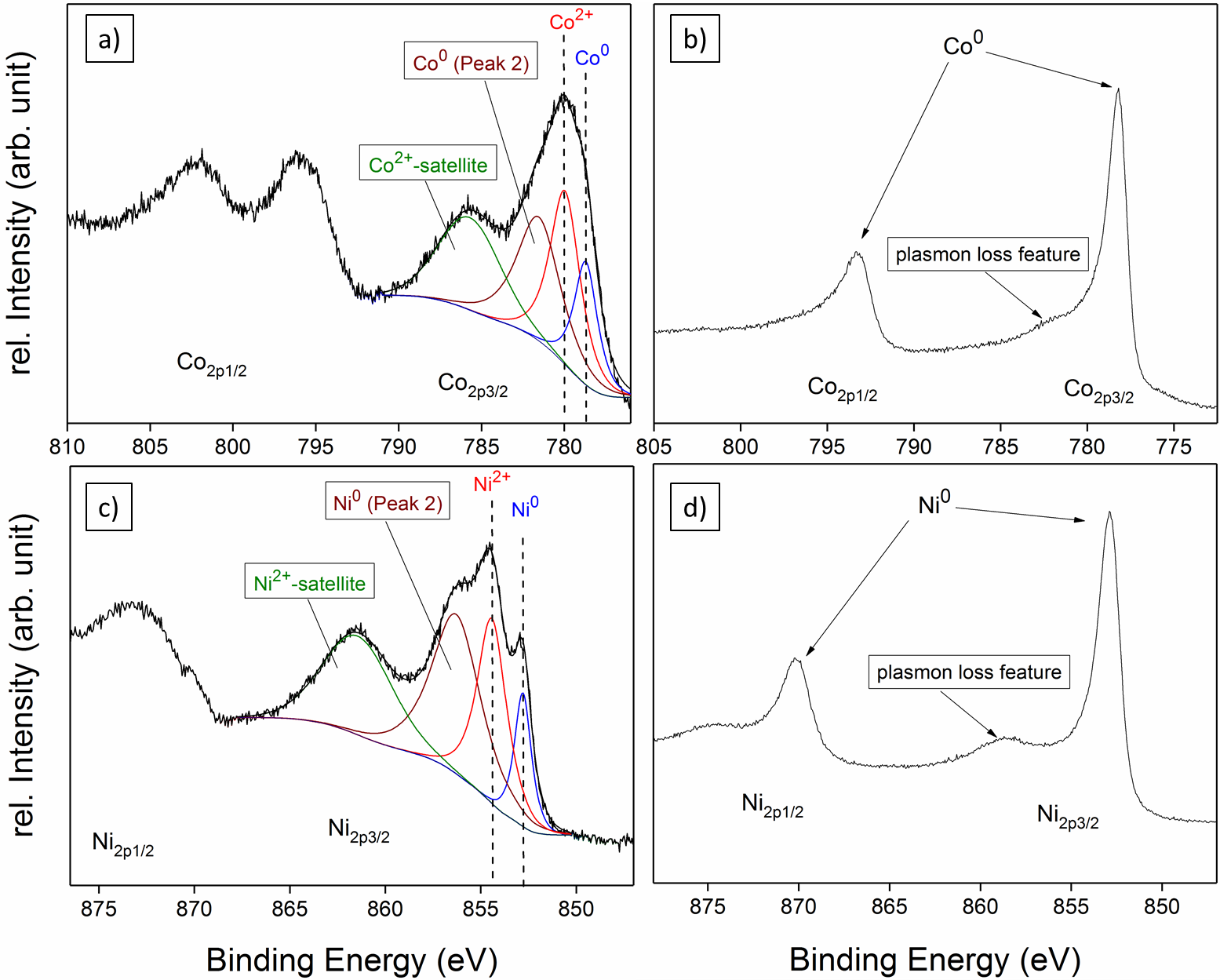
**FIG. S1**: a) High-resolution N 1s XPS spectrum after one precursor pulse using [Co(tfb-dmpda)2] **4b**, b) high-resolution O 1s XPS spectrum of the free ligand Htfb-dmpda **3b** deposited on a Si-wafer.



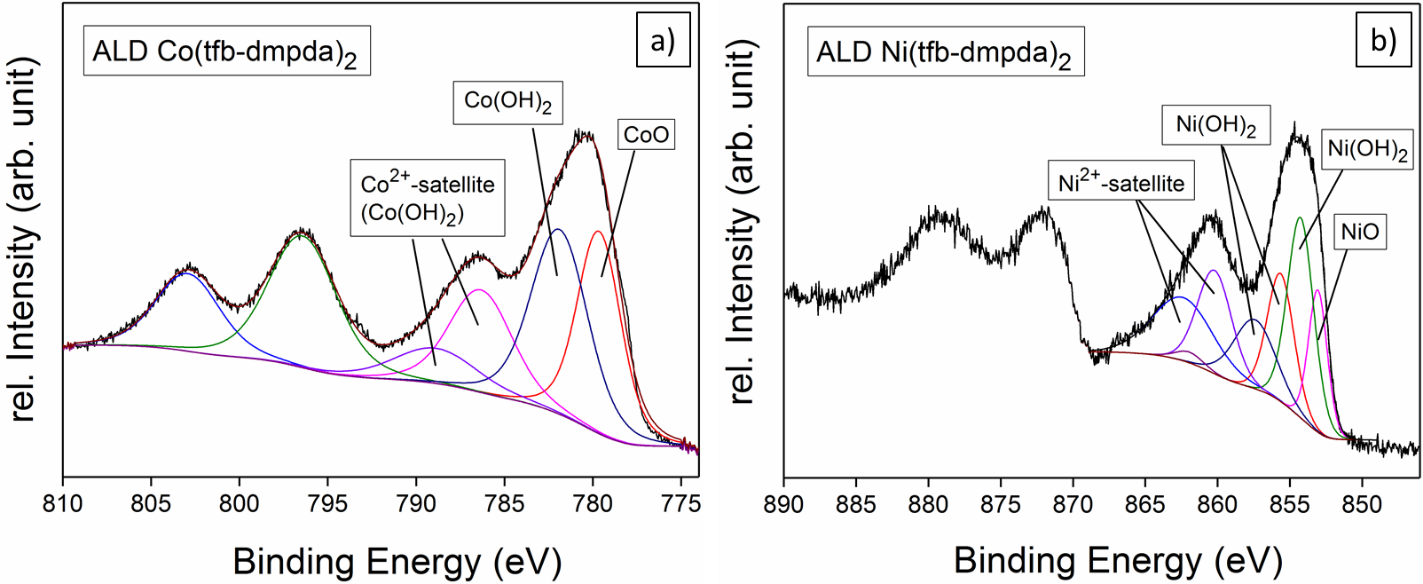
**FIG. S2:** Cross-section SEM analysis: a) ALD-deposited and hydrogen plasma treated cobalt film with a thickness of 260 nm (on a Si-wafer). b) Nickel film deposited under similar conditions with a thickness of 200 nm.



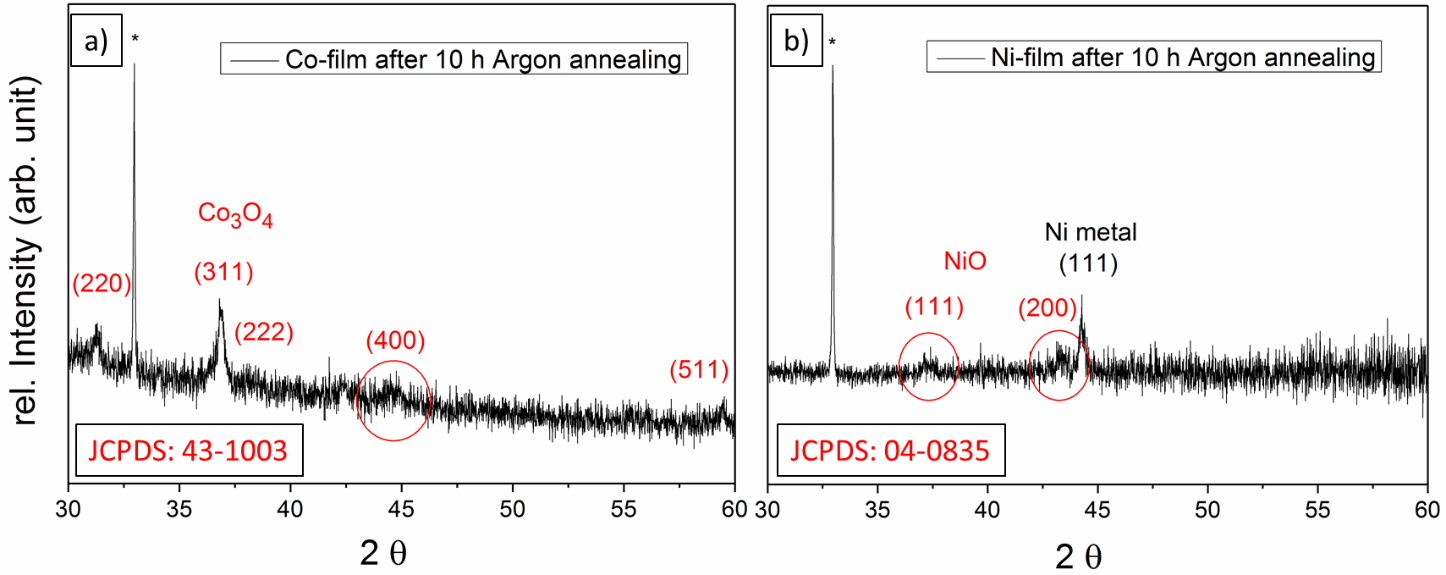
**FIG. S3:** AFM micrographs of a) metallic cobalt after H2-plasma treatment, b) metallic nickel after H2-plasma treatment.



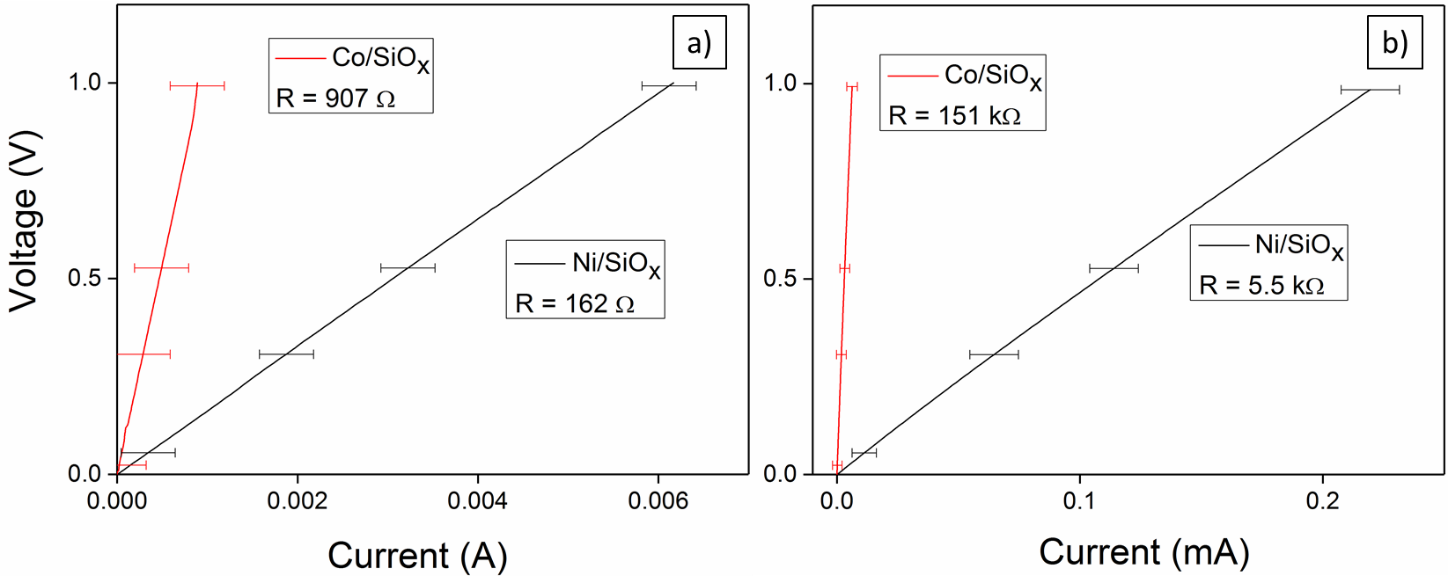
**FIG. S4:** High-resolution Co 2p XPS spectra of plasma treated cobalt films a) before and b) after argon etching, high-resolution Ni 2p XPS spectra of plasma treated nickel films c) before and d) after argon etching.



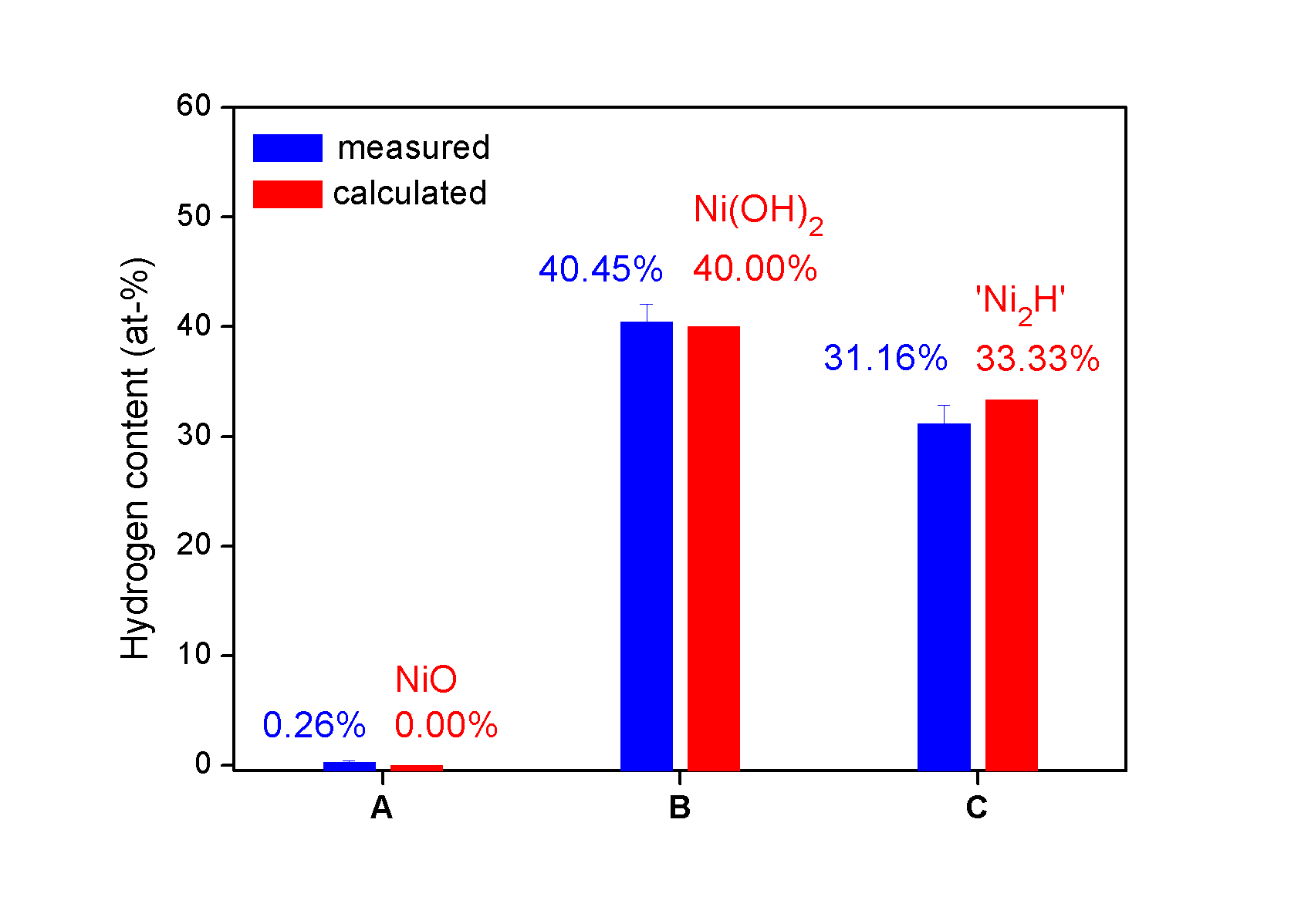
**FIG. S5:** High-resolution a) Co 2p XPS spectrum of as deposited Co films using [Co(tfb-dmpda)2] **4b** without post-deposition H2-treatment and b) Ni 2p XPS spectrum of as deposited Ni films using [Ni(tfb-dmpda)2] **5b** without post-deposition H2-treatment, subjected to argon sputtering.



**FIG. S6:** XRD analysis of a) cobalt and b) nickel films after additional annealing under argon atmosphere deposited on Si-wafers (\*).



**FIG. S7:** Resistance measurements of cobalt and nickel films after a) 500 cycles and b) 1000 cycles of post-deposition hydrogen radical treatment.



**FIG. S8**: Nuclear reaction analysis to quantify the hydrogen content in the following samples, A: deposition of NiO using oxygen plasma, B: deposition of Ni(OH)2 using hydrogen plasma as activant, C: B with excessive hydrogen plasma as etchant presumably forming nickel hydride.