

Synthesis and characterization of polymer-derived anti-perovskite nitride/amorphous silicon nitride nanocomposites

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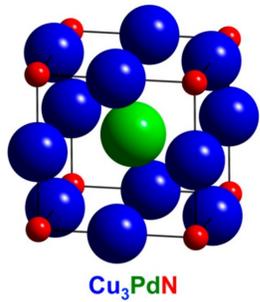
Introduction

Final target:

Synthesis of Cu_3PdN anti-perovskite nitride/amorphous silicon nitride (a-SiN)

Compared to Cu_3N and Pd, Cu_3PdN displays:

- superior ORR activity (Fuel Cell)
 - higher mass activity
 - better stability
- => Catalytic properties



The synthesis of Cu_3PdN nanoparticles is highly challenging but a required step developing catalytic properties for applications in clean energy and sustainable development.

First step: Investigate the formation of Pd/a-SiN and Cu/a-SiN

Second step: Utilize the new knowledge gained in the first step to perform the synthesis of $\text{Cu}_3\text{PdN/a-SiN}$

Elaboration Process: The Polymer-Derived Ceramics (PDCs) route

PDCs route

Preformed polymer (PHPS)

Chemical modification
(MCl_x , M = Pd and/or Cu)

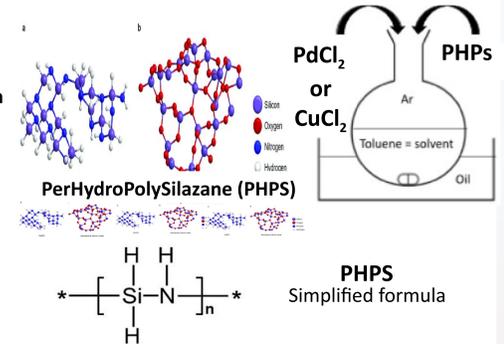
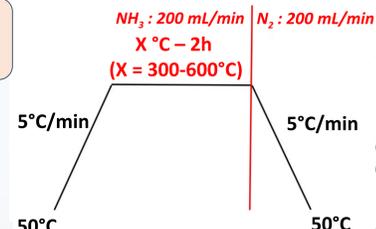
Preceramic polymer

Pyrolysis

Nanocomposite

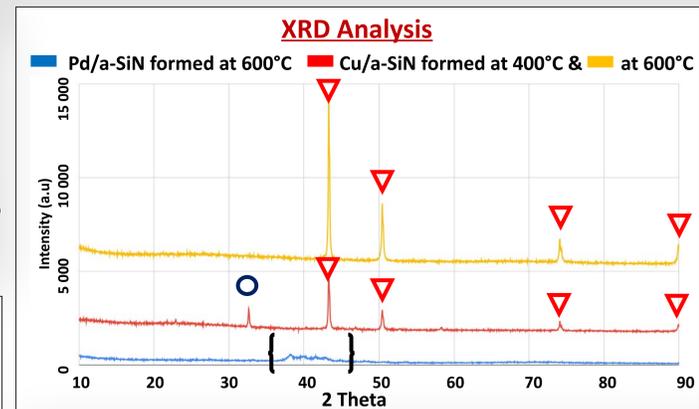
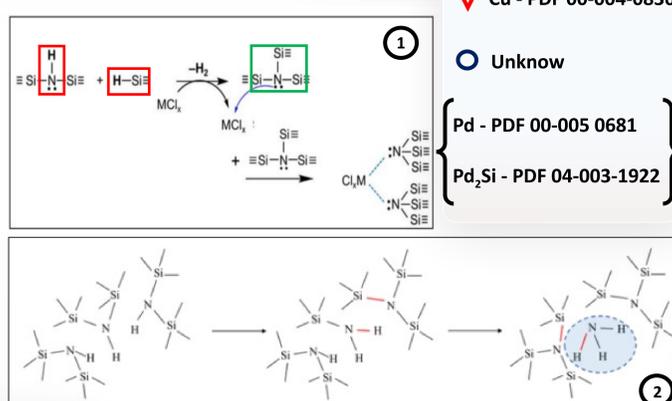
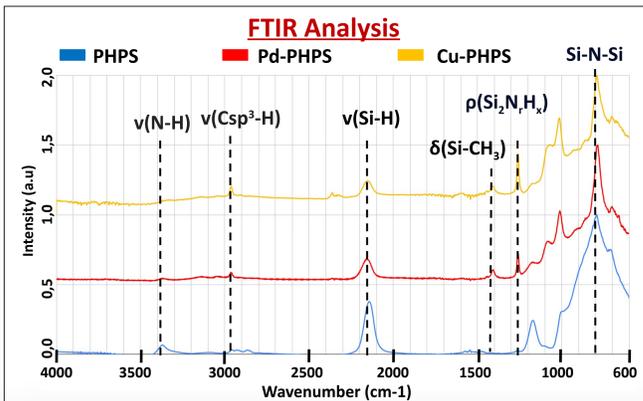
Made of metal-based NPs
in-situ formed in the a-SiN
matrix upon pyrolysis

- *High purity
- *Control of the microstructure
- *Additive-free synthesis
- *Low temperature ($T_{\text{max}} \approx 1200^\circ\text{C}$)
- *Shaping potential

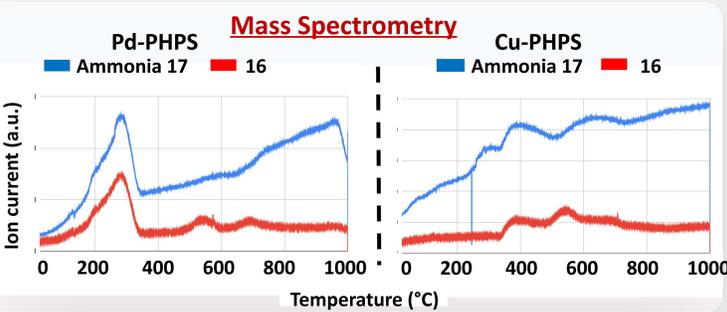


Results and Discussion

First step:

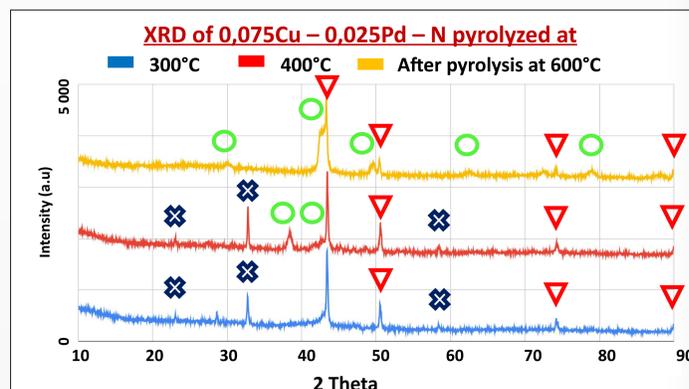
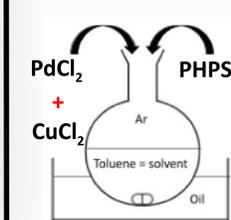


- We obtained the pure Pd and Cu phases as expected



- The decrease in the N-H and Si-H peaks, along with the increase in Si-N-Si => Dehydrogenation reaction ①
- The decrease in the N-H peak => Transamination reaction and the presence of ammonia in MS supports this mechanism ②

Second step:



- ⊗ $\text{Cu}_3\text{Pd}_{0.985}\text{N}$ - PDF 01-080-1885
 - ▽ Cu - PDF 00-004-0836
 - Pd_2Si - PDF - 04-003-1922
- Isolation of the Cu_3PdN antiperovskite phase at 300 & 400°C!
- Pyrolysis at 600°C eliminates this phase
- Presence of Cu and Pd_2Si at 400°C

Conclusion and Perspectives

- ✓ FTIR analysis identified two reaction (dehydrogenation and transamination) and MS confirmed the transamination reaction
- ✓ Presence of Pd and Cu phase in XRD respectively for Pd-PHPS and Cu-PHPS and formation of Cu_3PdN in the Second step
- ✗ Presence of Pd_2Si in Pd-PHPS XRD, an unknown peak appears in Cu-PHPS XRD at 400°C and presence of secondary phases in the Second step

To go further:

- Try further reducing the pyrolysis temperature and extending its duration to assess the impact of both parameters on the formation of Cu_3PdN
- Conduct SEM observations and EDS mapping to examine the distribution of elements. This will help determine whether the Cu phase observed in XRD results from a homogenization issue
- Extend the concept towards other anti-perovskite nitride & carbide phases for clean energy > JSPS scholarship of Dr. Maxime Cheype