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Coordination complexes with multi electron transfers as ion-battery electrodes

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Introduction

Context : Today, due to the continuous increase of energy storage devices (mobile and stationary), research dedicated to battery is still a main challenge. Li-ion technology, leader in this field, involves efficient but limited electrode materials resulting in the development of new materials.

Classical Li-ion
batteries electrode
materials



- ✓ Unsolubility, good cycling
- ✓ High energy density
- ✗ Not versatile, expensive, toxic
- ✗ Geostrategic resources
- ✗ Limited power and speed of charge

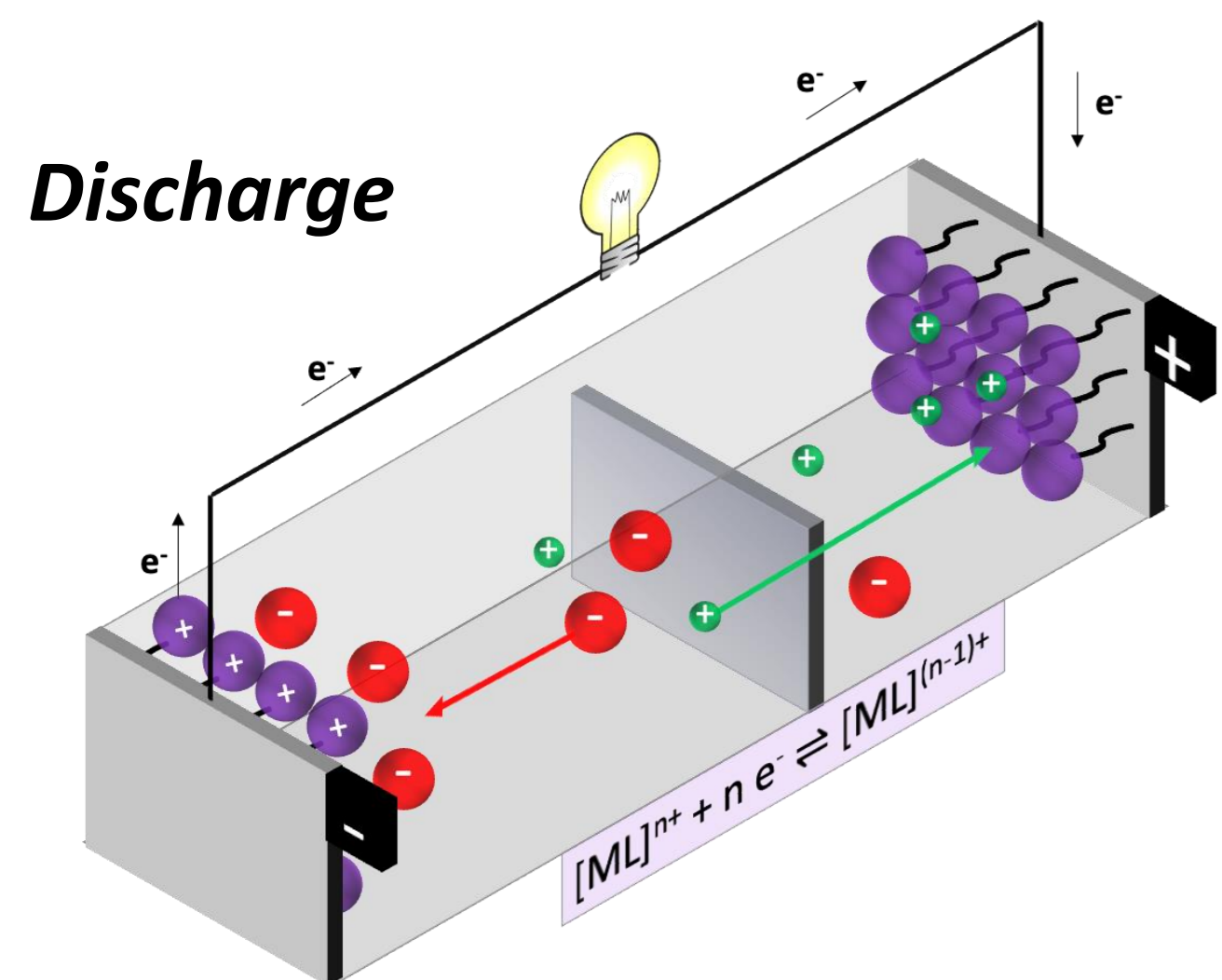
- ✓ Versatility
- ✓ Unsolubility, reversible redox properties
- ✓ High capacity (C_{th}), oxidation potential
- ✓ Fast electron transfer (k_0)
- ✓ Low cost, low toxicity

Coordination
complexes polymers
as electrode



Objectives, Strategy and Materials

Coordination polymers as **redox active** electrodes materials



Key material: fast charge, high power, beyond Li-ion

- : coordination polymers
- : cations (Li^+ , Na^+ , K^+ , ...)
- : anions (ClO_4^- , $TFSI^-$, ...)

Challenge : Highest capacity as possible :

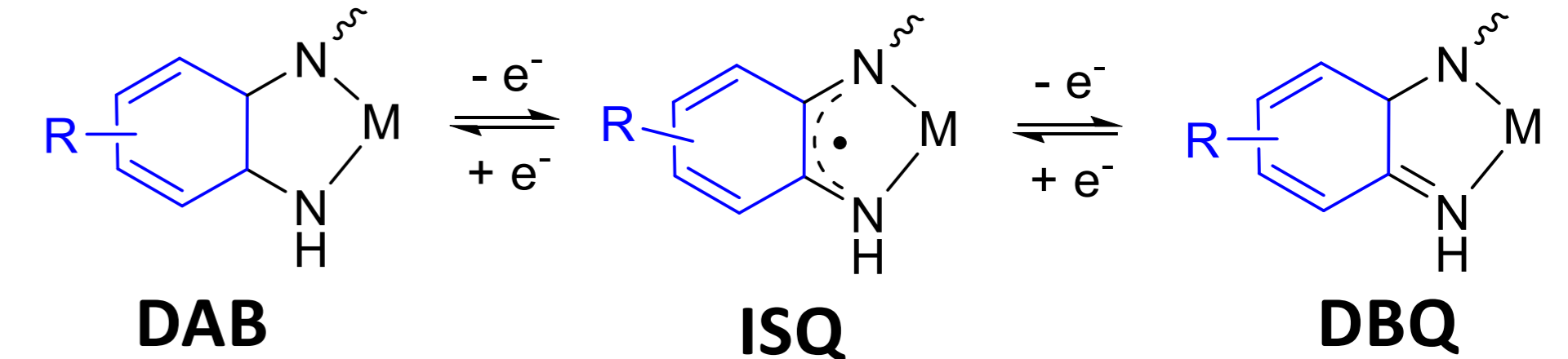
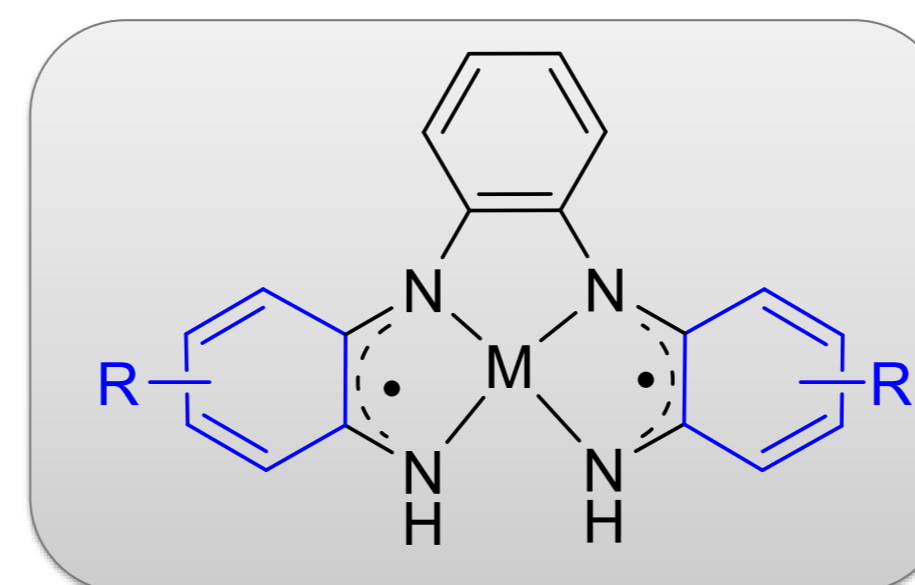
- Multiple electrons systems ($n_e^- \uparrow$)
- Light compounds ($M_a \downarrow$)

$$C_{th} = \frac{m_a \times F}{M_a} n_e^-$$

Optimization : design at the **molecular scale**

Structure of interest: tetradentate complexes based on o-phenylenediamine¹

- A redox active ligand : reversible monoelectronic exchanges

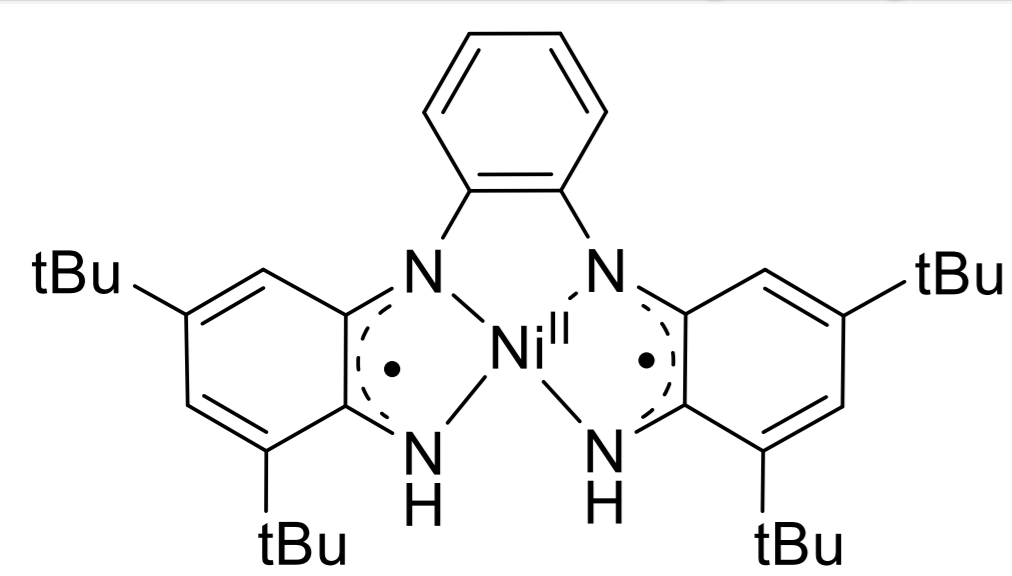


- 2 ISQ units per complex ($n_e^- \uparrow$)

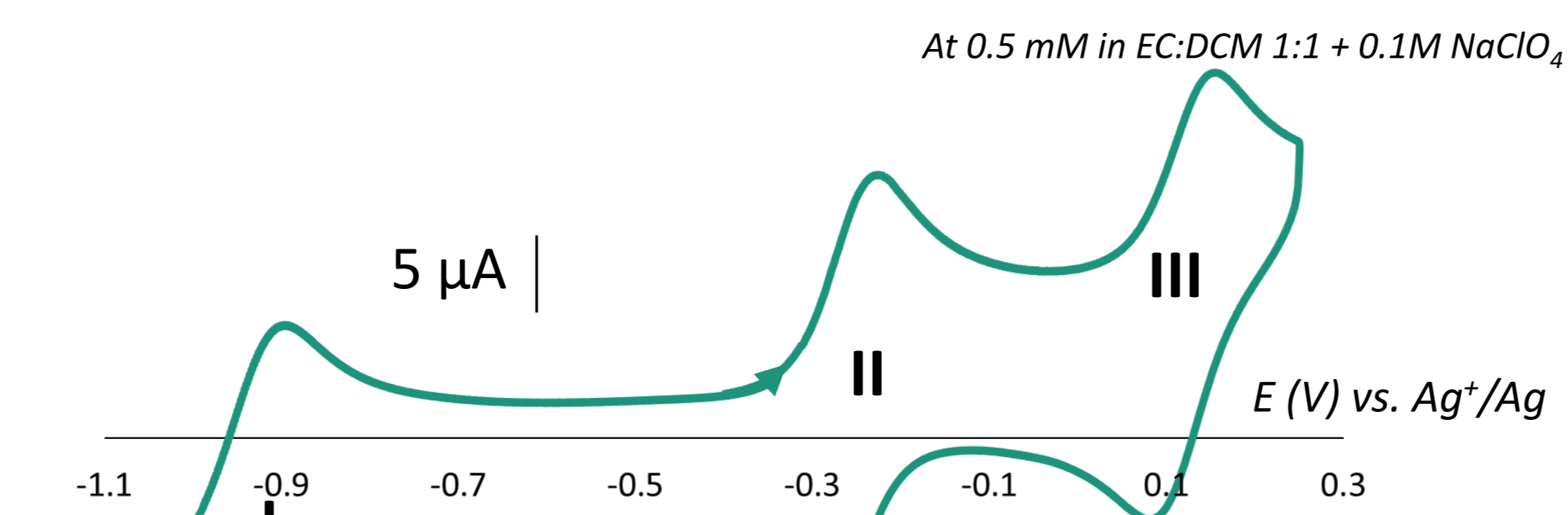
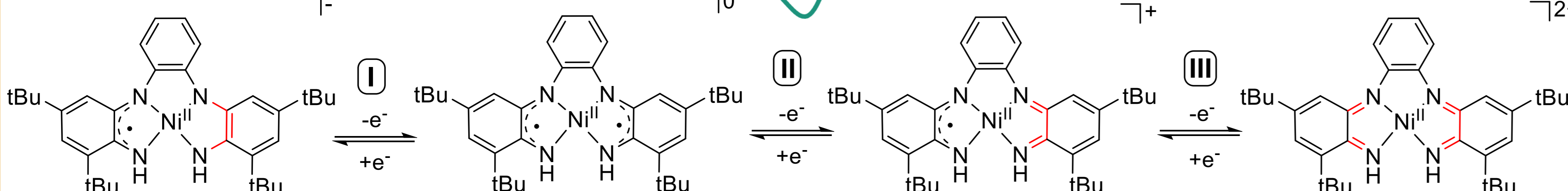
Reach until **4 e⁻ transfers : $C_{th} \times 4!$ ($> 250 \text{ mA.h.g}^{-1}$)**

- **R** : modulation of **potential** and **molar mass** ($M_a \downarrow$)

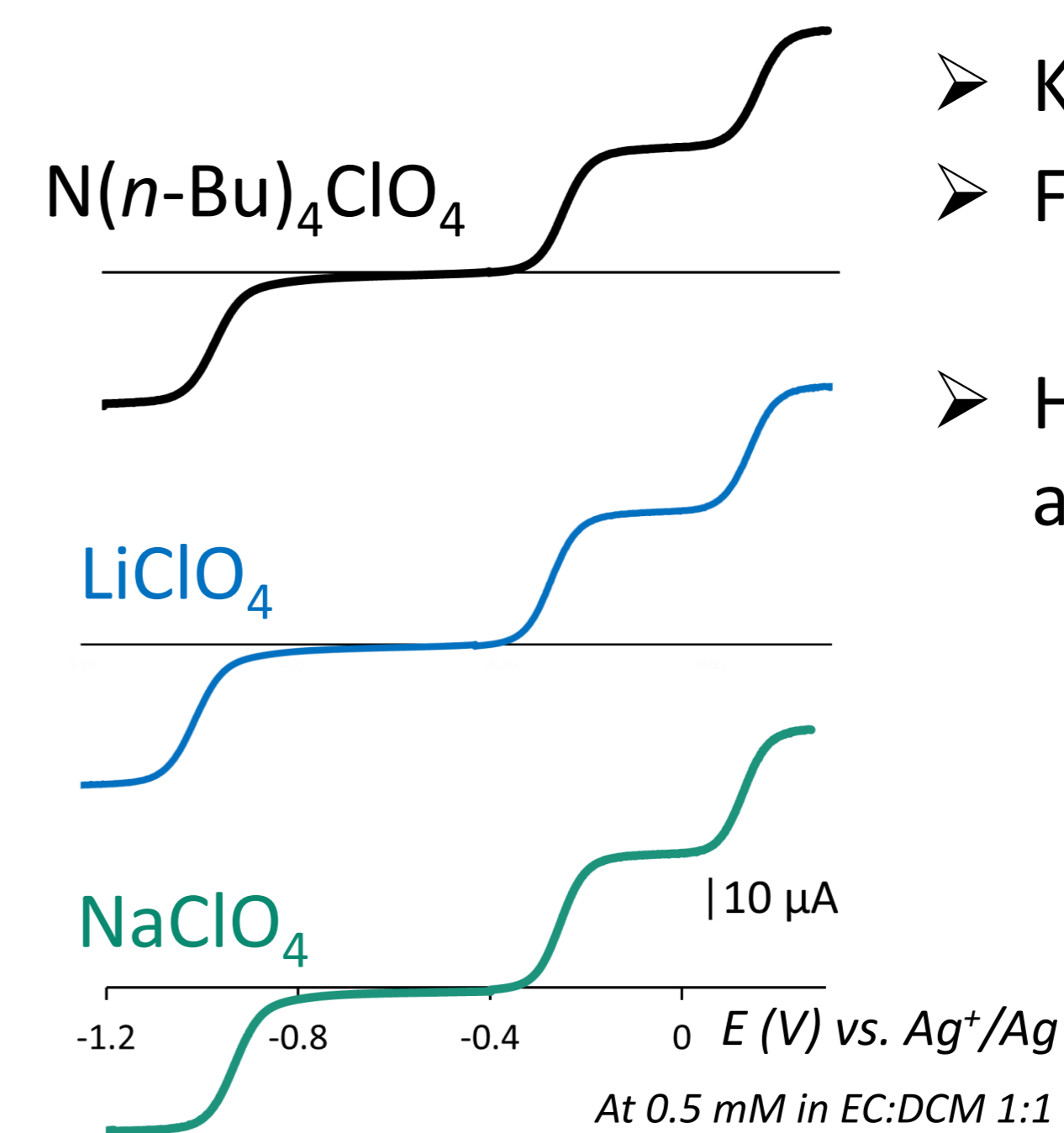
Electrochemical properties



- Competitive² $C_{th} = 147 \text{ mA.h.g}^{-1}$
- **3 electrons** exchanged
- **Reversible** multi step redox systems



Versatility in electrolytes

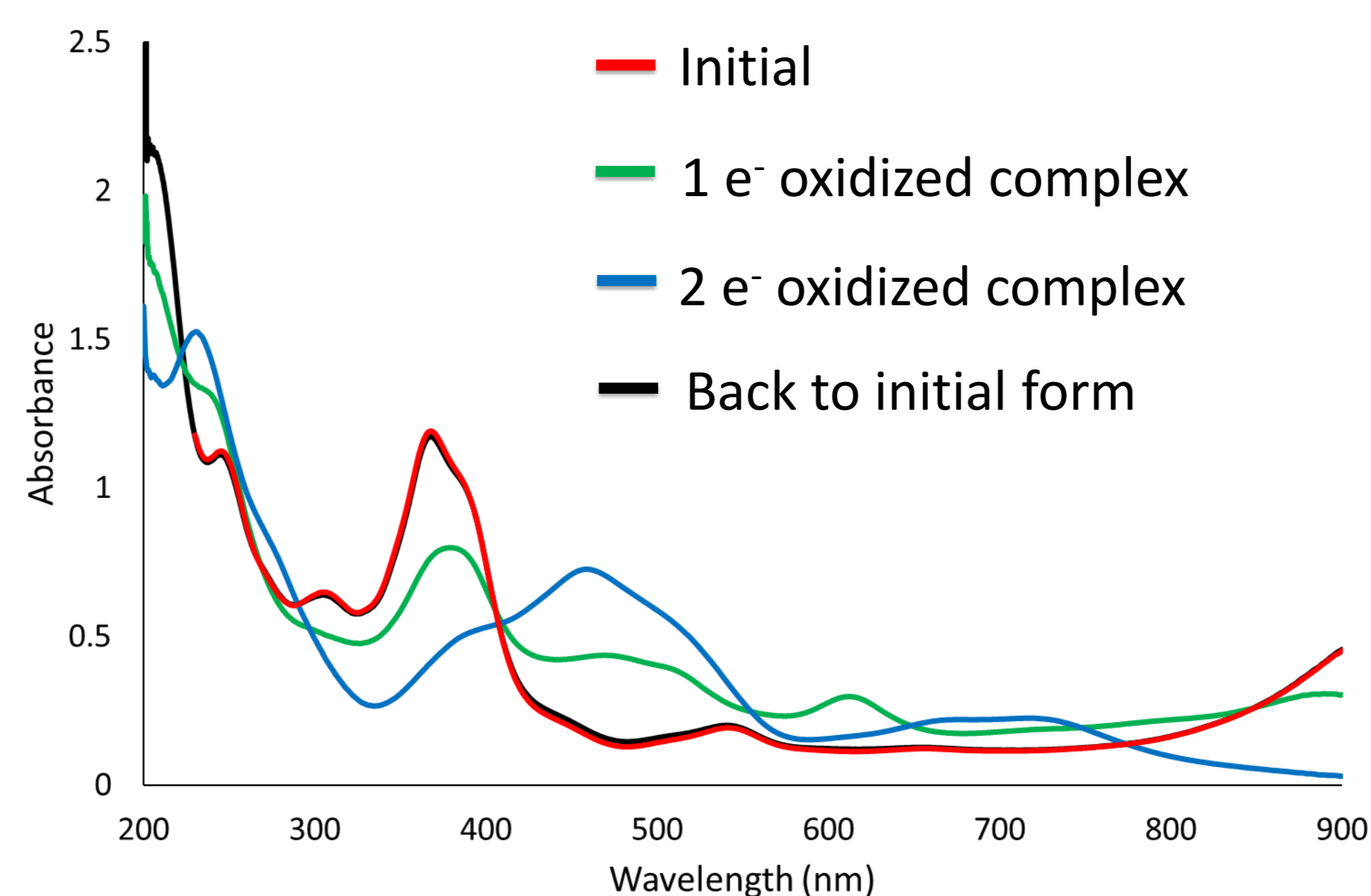
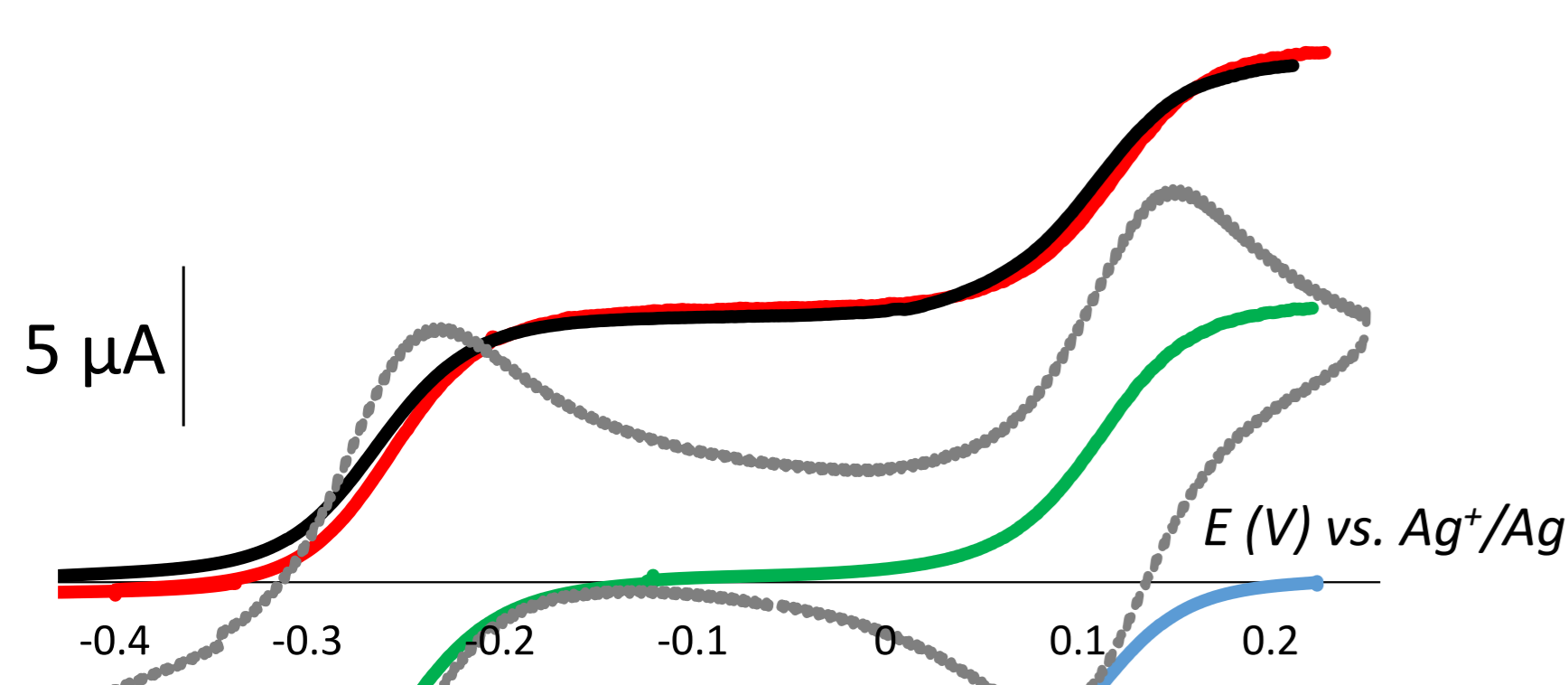


- Keeping the **3 systems**
- Fast electron transfers $k_0 \sim 10^{-2} \text{ cm.s}^{-1}$
- High C rates could be achieved

Versatility
Fast kinetics
High power batteries

Spectro-electrochemical investigations

- 3 electrodes setup : WE (Glassy Carbon), CE (Pt), Ref (Ag)
- **Electrolysis** at - 0.15 (**1 e⁻**), 0.25 (**2 e⁻**) and - 0.45 V vs. Ag^+/Ag (**back**)

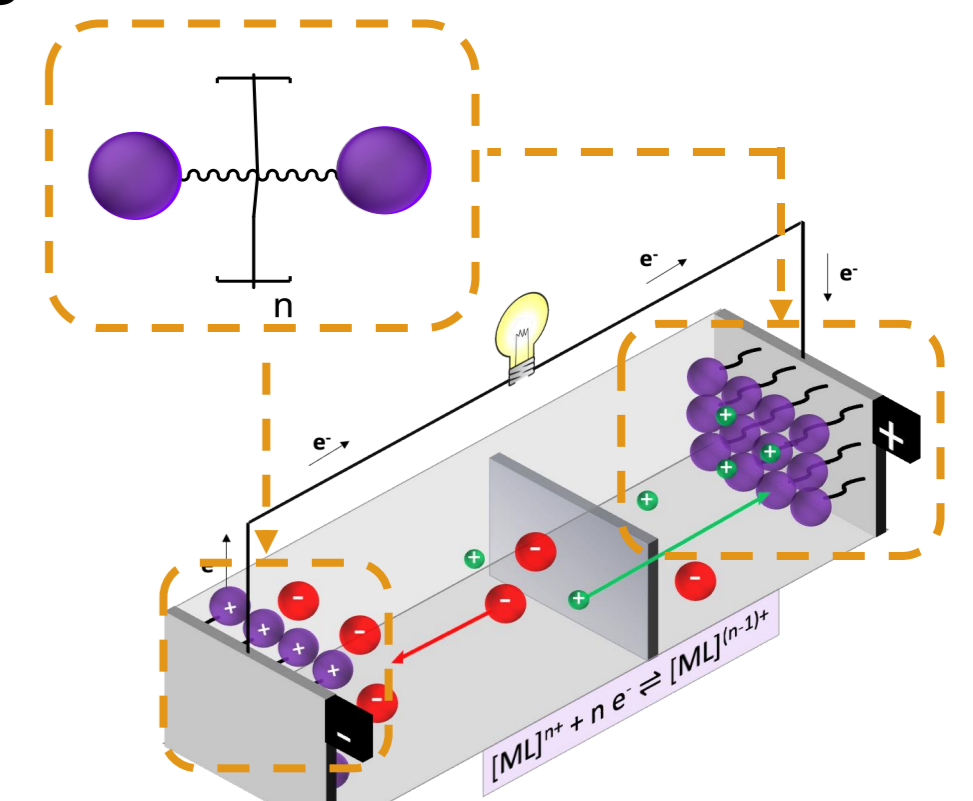
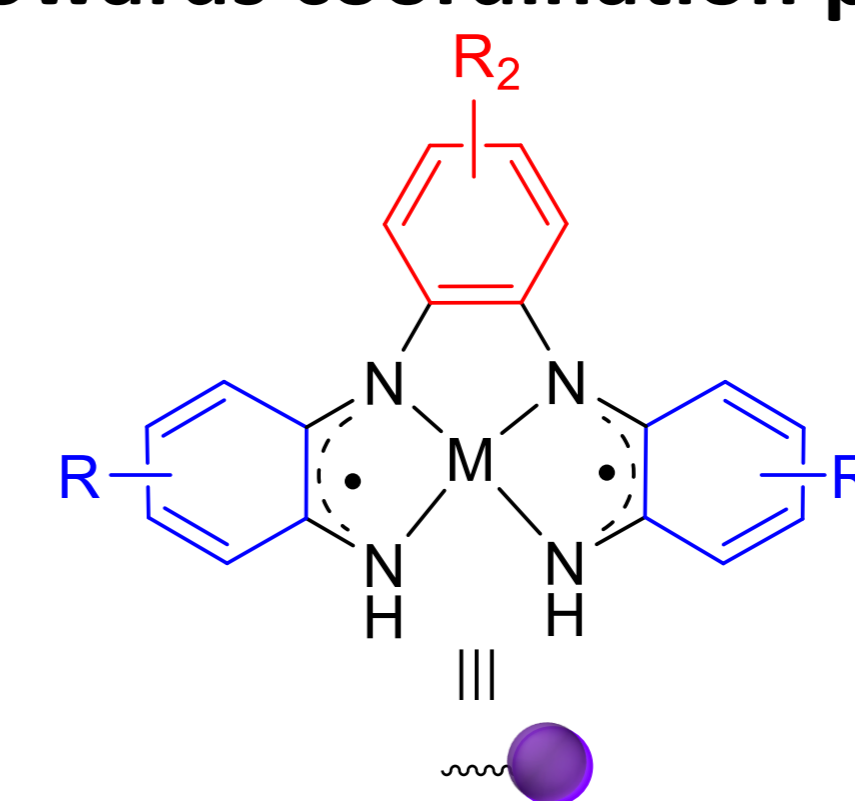


- Fully **reversible** systems
- **Stable** during 'long' time experiment
- Superimposable 'initial' and 'back' curves
- No parasitic reaction

Ideal for battery application

Ongoing work

Towards coordination polymers



- **R₂** : anchoring group for **polymer integration**
- **Insoluble** electrode material in the electrolyte
- **Electrochemistry** of the polymer

Conclusion

- Complexes with **multi electron transfers**, **fast kinetics**, **high stability** and **reversibility**
- Allow to obtain **high power** and **fast charge** batteries
- Good candidate for **electrode material**