

# Interphasial and Thermal Reactivity Investigation of Silicon-Graphite Composite Anode Electrodes

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## Abstract

To meet the requirements of next-generation high energy density lithium-ion batteries (LIBs), Silicon (Si) and its derivatives (e.g. Si/C, Si rich  $\text{SiO}_x$ ) have gained huge attention as anode materials. However, despite the beneficial features (e.g. low cost and high capacity), Si shows limited cycle life, huge volume changes during (de-)alloying, low coulombic efficiency (CE), and unstable solid electrolyte interface (SEI). One of the most efficient, scalable, and economic strategy to overcome these problems is to modify the chemical composition and features of the SEI layer with the help of small doses of electrolyte additives.

## Conclusion

The addition of  $-\text{C}\equiv\text{N}$  functionalized TEOS to FEC/VC:

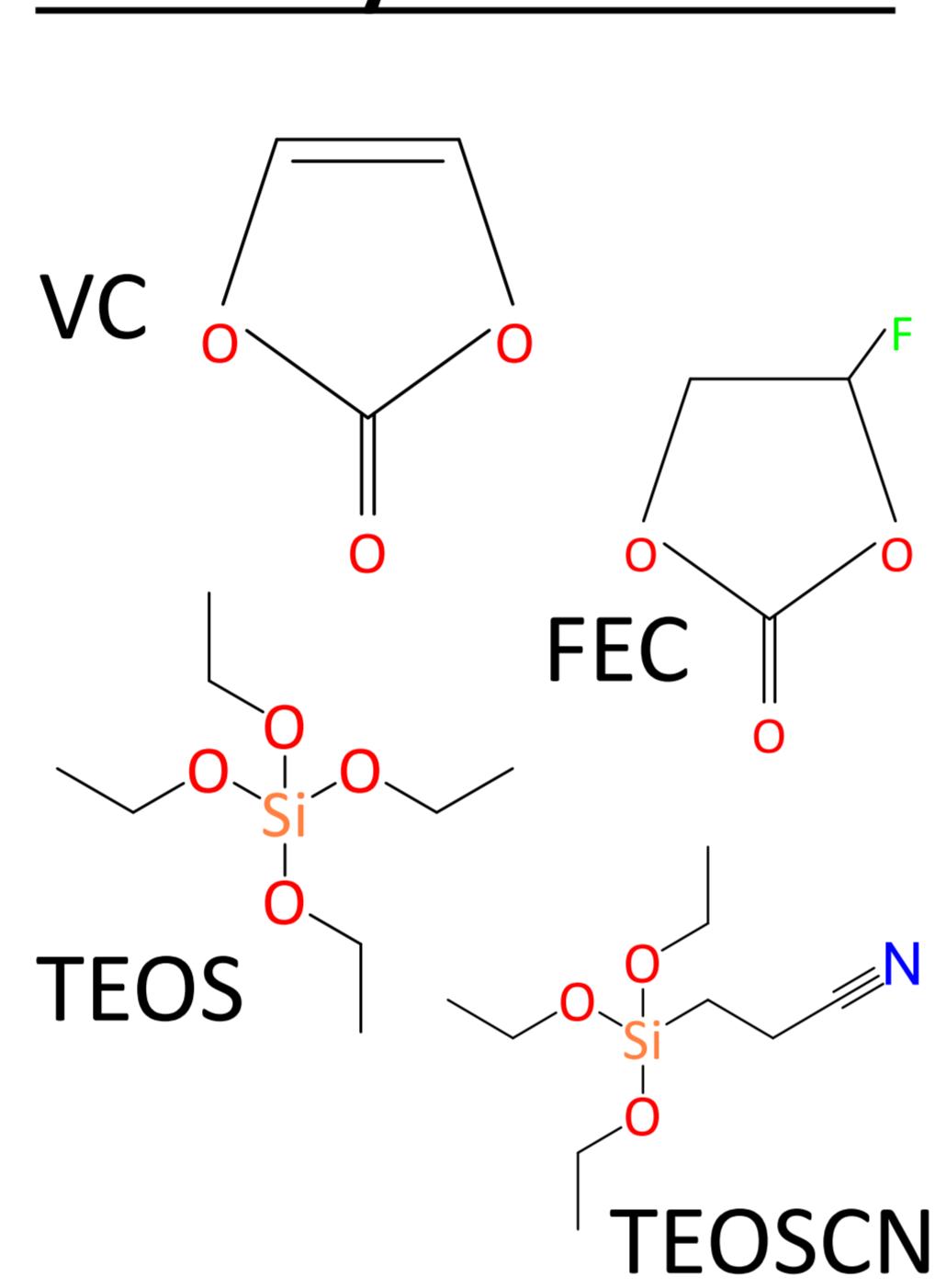
- Improves the electrochemical performance in terms of cyclability, capacity retention, and coulombic efficiency at 45°C
- Leads to the formation of an inorganic rich (e.g. LiF), thinner, and mechanically stable SEI layer
- Significantly lowers the heat of enthalpy while increasing the onset temperature for SEI decomposition of the main peak

## Experimental Details

### Pouch Cell Data

- Anode: Graphite/Si-alloy
- Cathode: NMC 622
- Base Electrolyte (BE): 1M  $\text{LiPF}_6$  EC:DEC with VC/FEC/(TEOSCN)
- Charge: CC 1C to 4.2V and CV until C/20
- Discharge: CC C/2 to 2.5V

### Electrolyte Additives



### XPS conditions

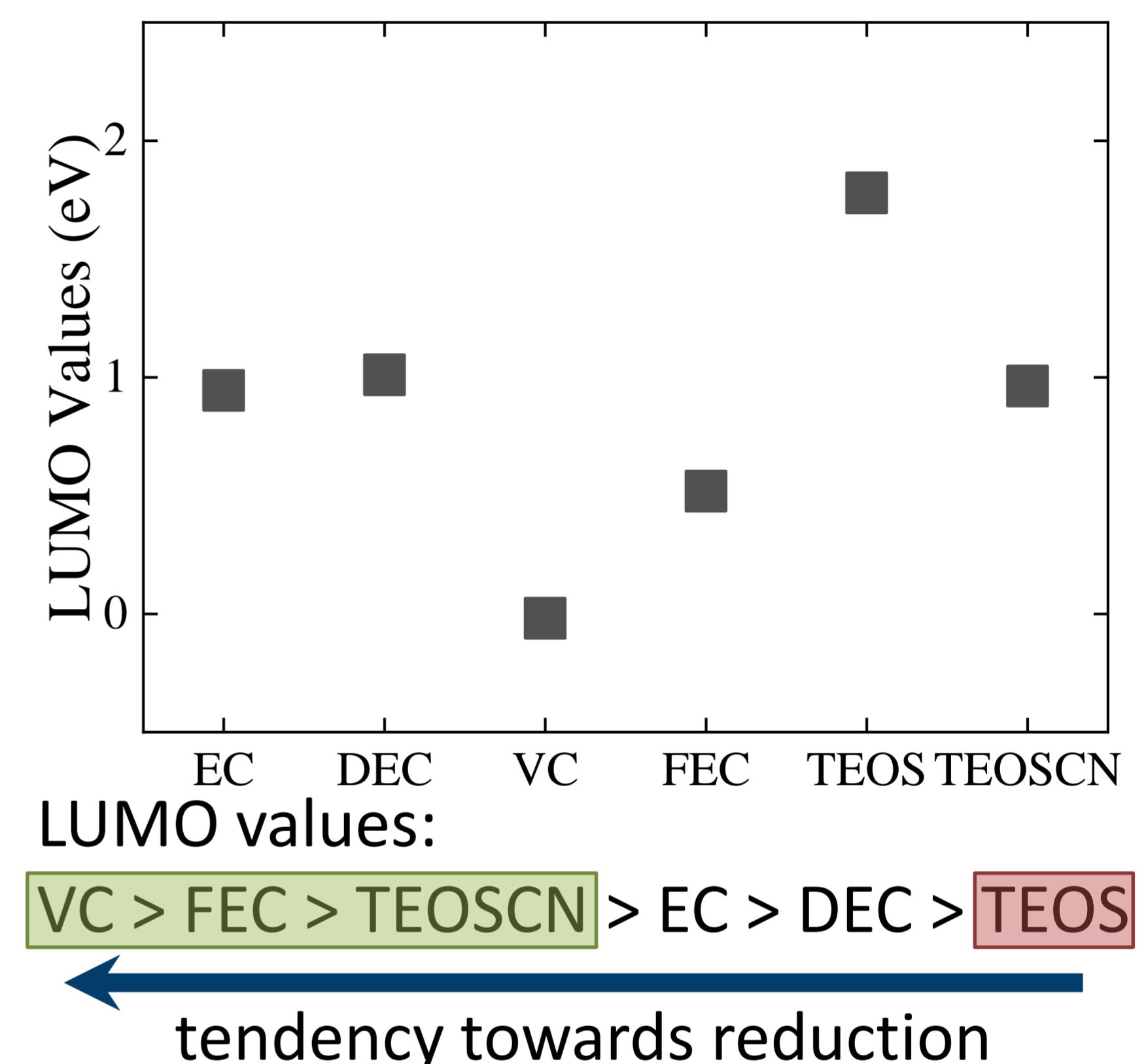
- $\text{AlK}_{\alpha}$  radiation (1486.3 eV)
- emission ( $0^\circ$ )
- pass energy (160 eV)

### DSC conditions

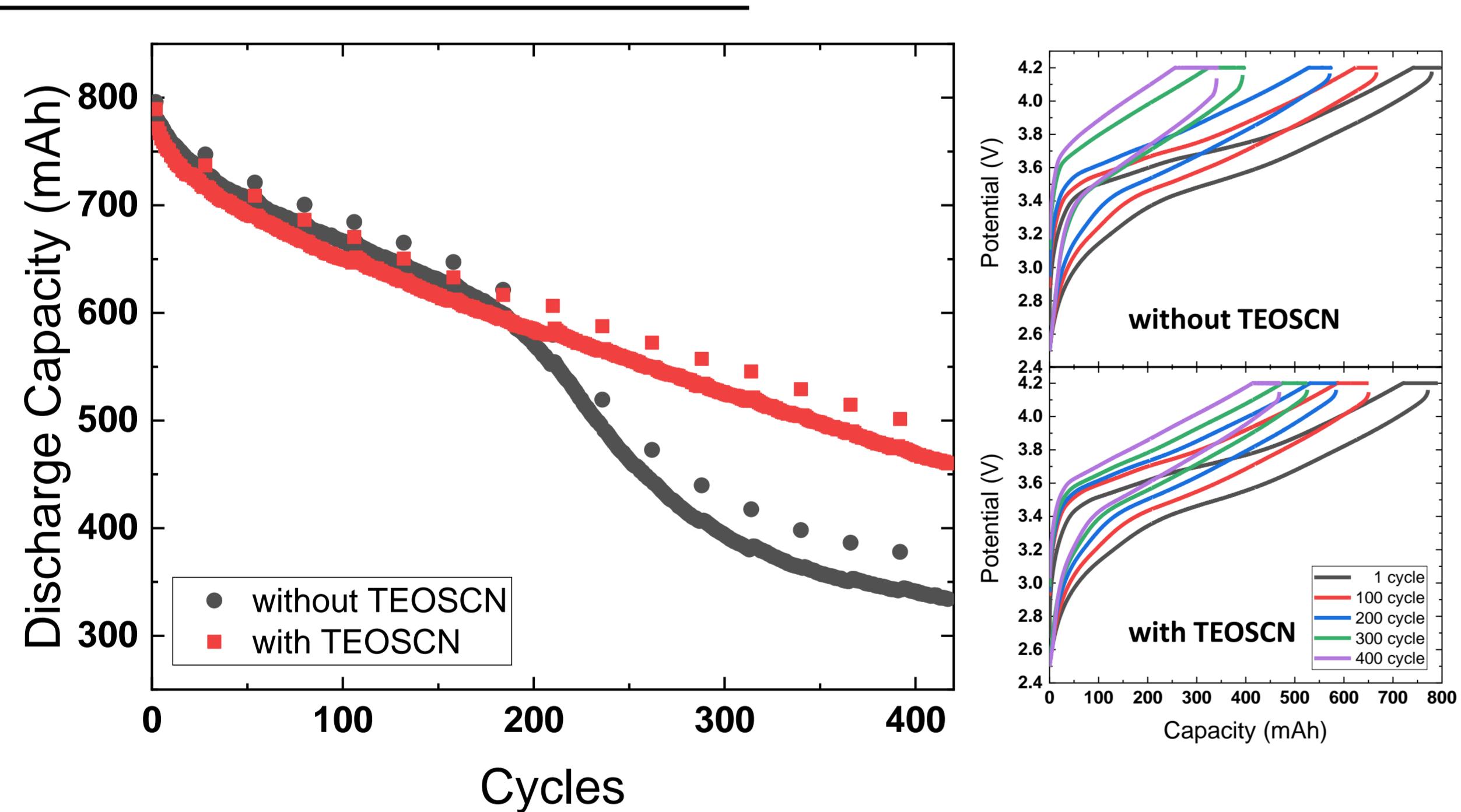
- heating rate ( $5 \text{ K} \cdot \text{min}^{-1}$ )
- temperature range ( $20^\circ\text{C} - 350^\circ\text{C}$ )
- $\text{N}_2$  atmosphere

## Results

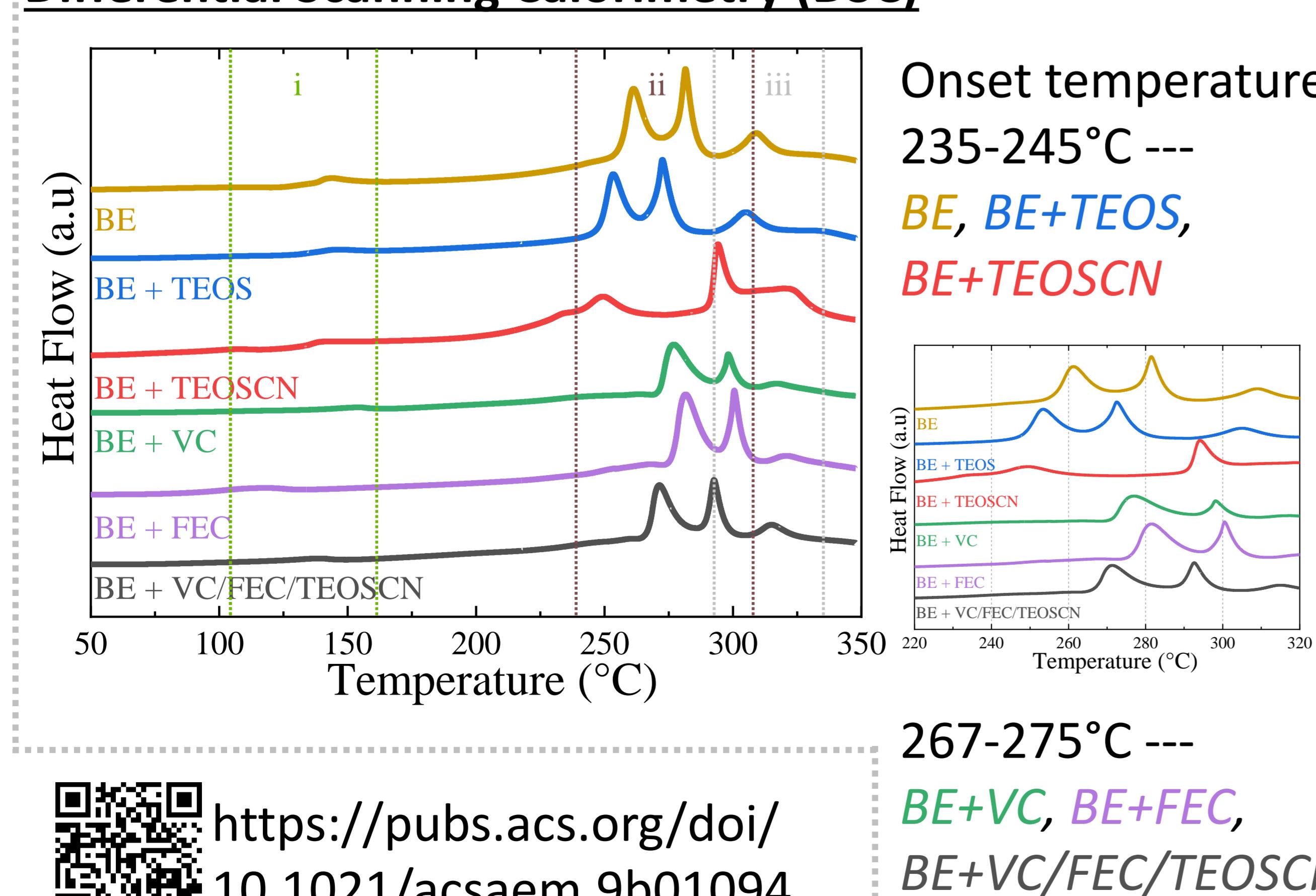
### Density Functional Theory (DFT)



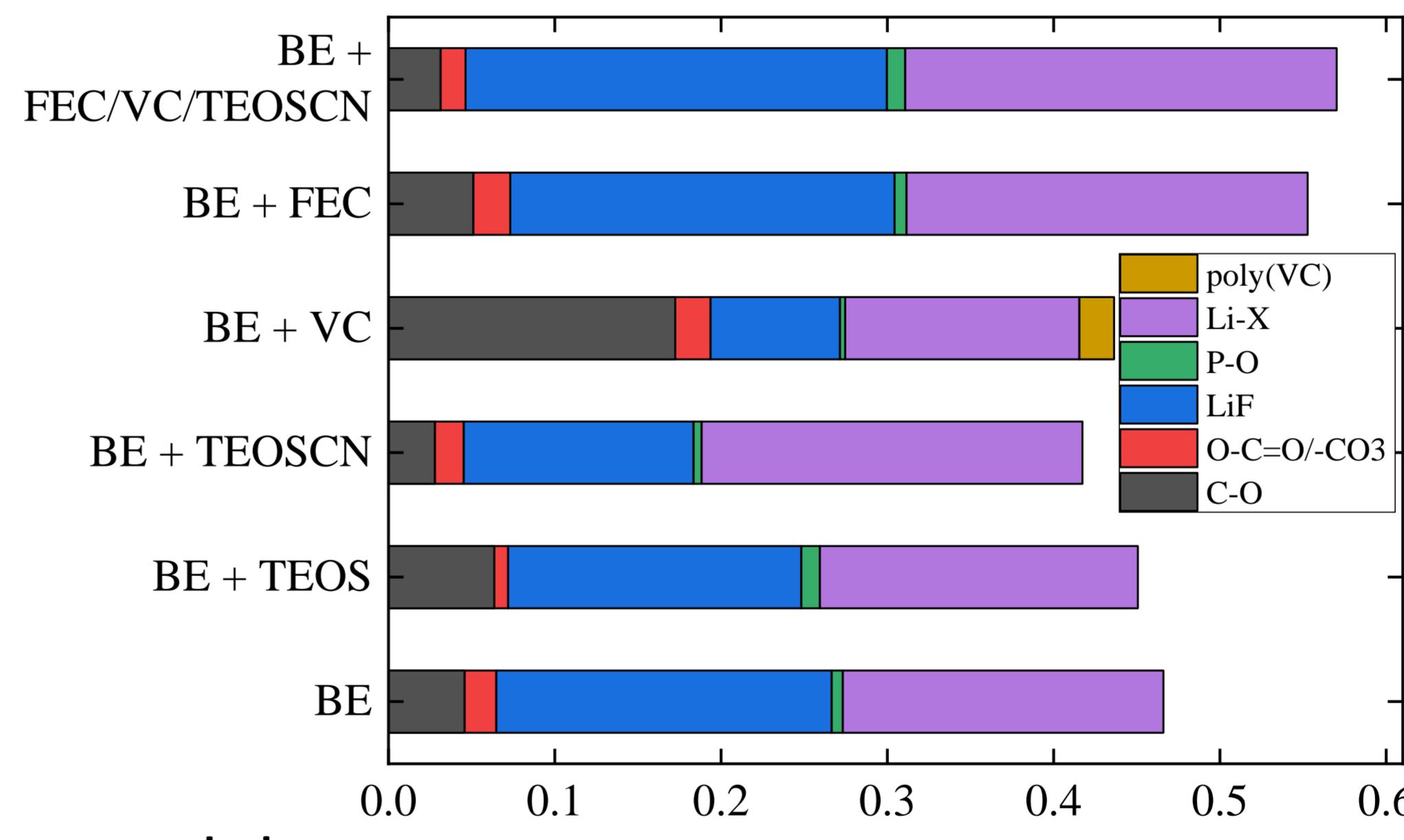
### Electrochemical Performance



### Differential Scanning Calorimetry (DSC)



### X-ray photoelectron spectroscopy (XPS): SEI Building Components

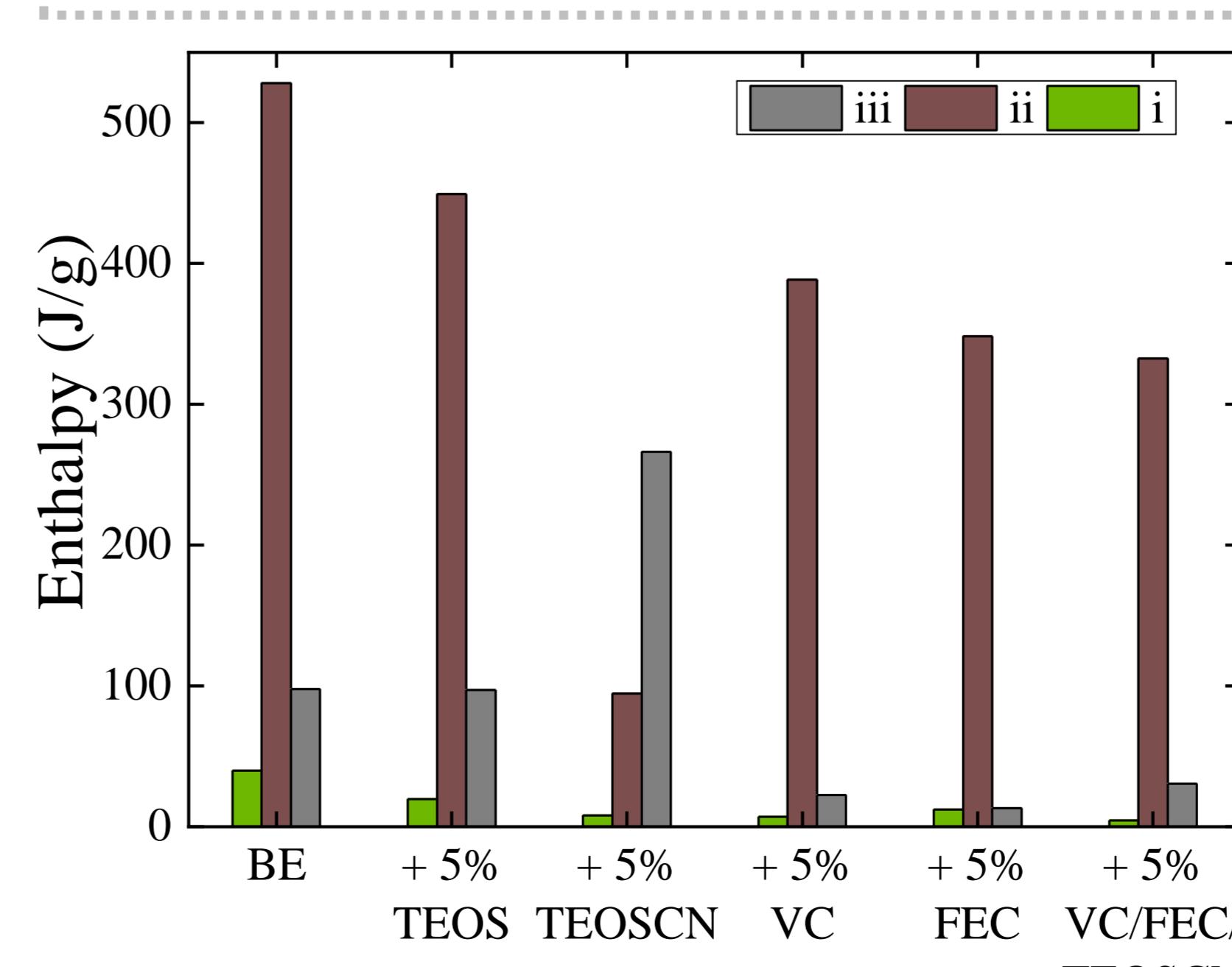


shear modulus:

Atomic fraction:  $\text{LiF} (55.14 \text{ GPa}) > \text{Li}_2\text{O} > \text{Li}_2\text{CO}_3 > \text{organic components} (<1 \text{ GPa})$ <sup>[1]</sup>

solubility in alkyl carbonates (EC and DEC):

organic components > LiF >  $(\text{LiCO}_2)_2$  >  $\text{Li}_2\text{CO}_3$  >  $\text{Li}_2\text{O}$ <sup>[1]</sup>



- Addition of electrolyte additives reduce total enthalpy
- $-\text{C}\equiv\text{N}$  functionalized TEOS significantly decrease the exothermic heat release of the most important peak