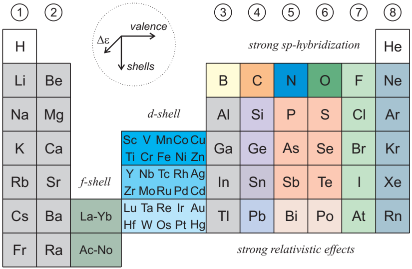
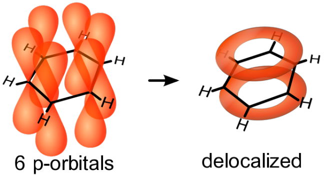
Intro 1/4: What is organic semiconductor

1. **Organic** – built from C with terminal H, possibly with isovalent substitutions (N for CH, O/S for CH_2, F/Cl for H)
2. π-**conjugated** – only π-electrons on frontier orbitals





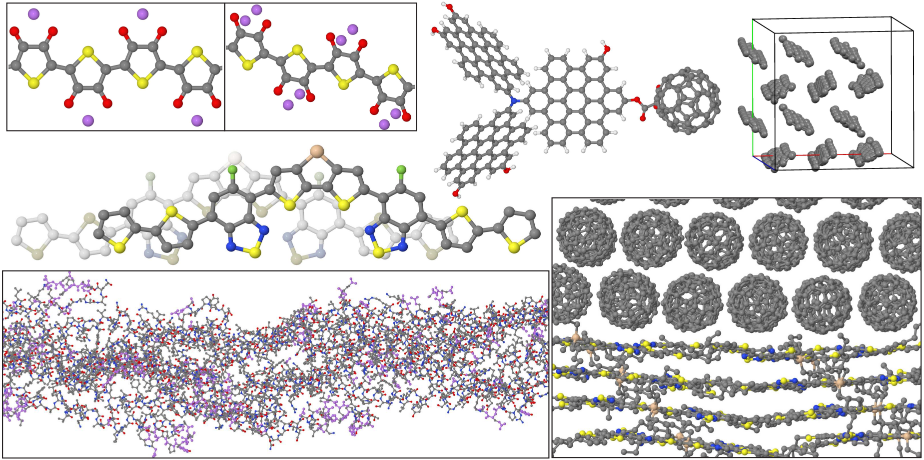
$$t_{sp2} = 3.26\frac{\hbar^2}{md^2} \gg t_{pp\pi} = 0.63\frac{\hbar^2}{md^2}$$
(Harrison's parameterization)

- Also **metal-organic** – include d-elements via $pd\pi$
- Also **hybrid** – include any electronic system in resonance

Number of organic semiconductors is comparable to number of inorganic ones
Display is an example of a competitive application

Intro 2/4: Structural diversity

From 0D to 3D, from rigid to soft, often heterogeneous with multiscale structuring



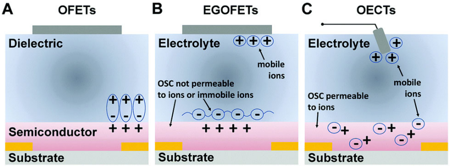
Intro 3/4: Some applications of organic semiconductors

Any electronic device can be made all-organic

Light emitters (most successful)

Adv Mater 33, 2005630 (2021)

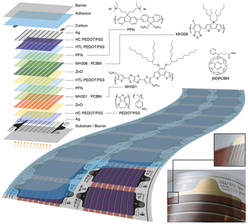
Transistors (most ubiquitous)



J Mater Chem C 6, 11778 (2018)

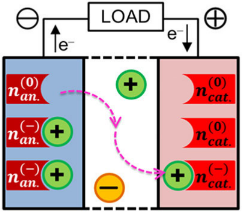
Solar cells (approaching 20% PCE)

Adv Energy Mater 11, 2002653 (2021)



Energy Environ Sci 7, 2925 (2014)

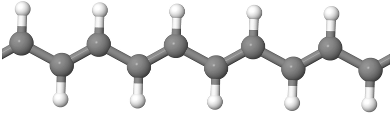
Batteries (early research)

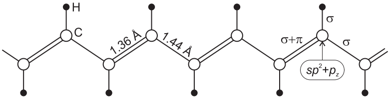


Chem Rev 120, 6490 (2020)

Intro 4/4: Challenges in modeling organic semiconductors

- Electron-electron correlations in low dimensional systems
  - Trans-polyacetylene (tPA) – 5 electrons in repeating unit – smallest real-world material for which majority of mainstream approaches fail





- Strong mode-specific electron-phonon coupling + soft structure
  - tPA – Peierls transition (C–C bond stretching mode) and solitons
  - Bond Length Alternation (BLA)
  - polaron formation
  - vibronic progression in spectra
- Scale gap problem – what is modeled is not what is measured