Nanoscale Designed Materials Solving Macroscale Problems:

PNNL's Nanoscience and Technology Initiative (NSTI)

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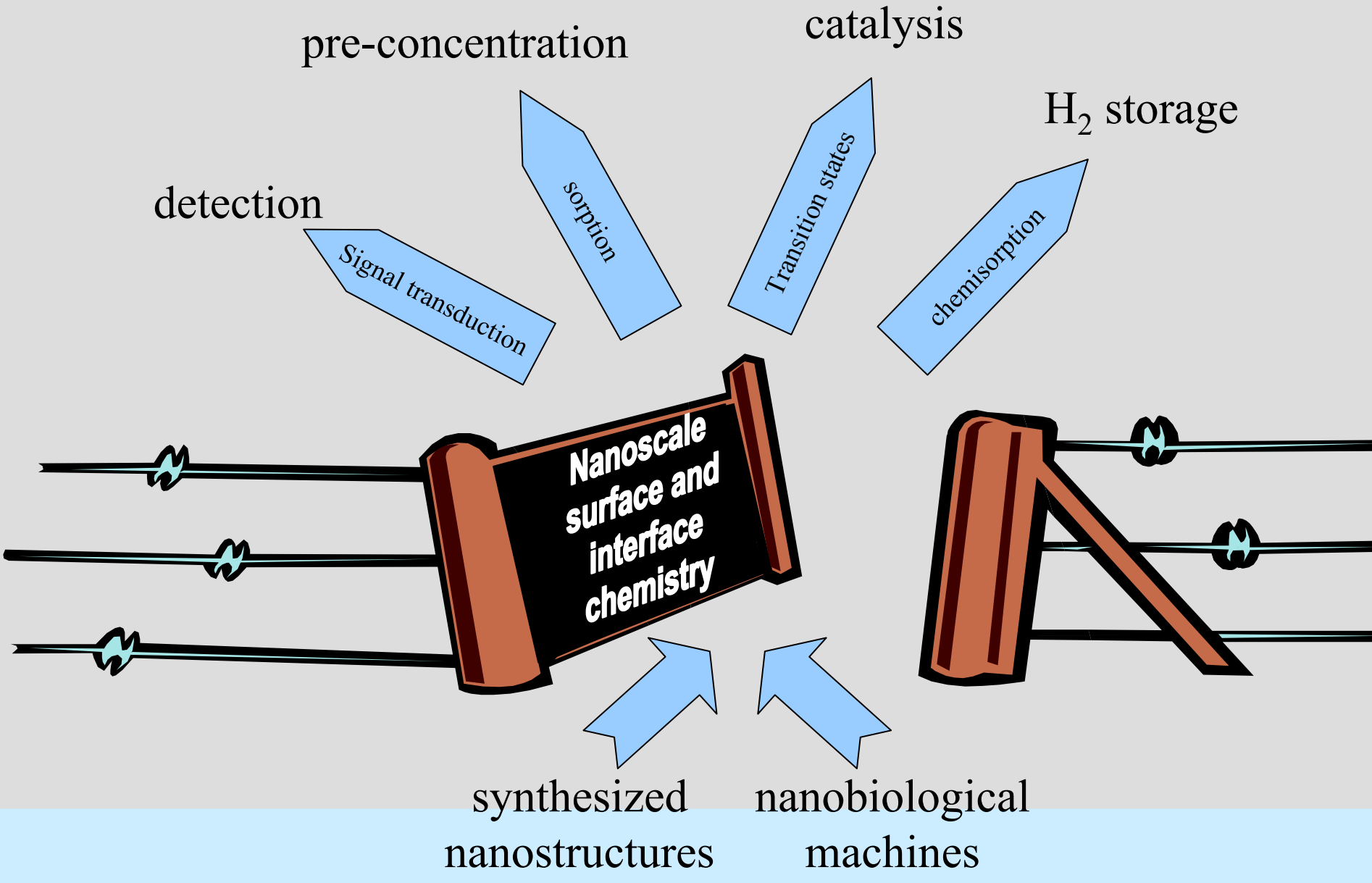
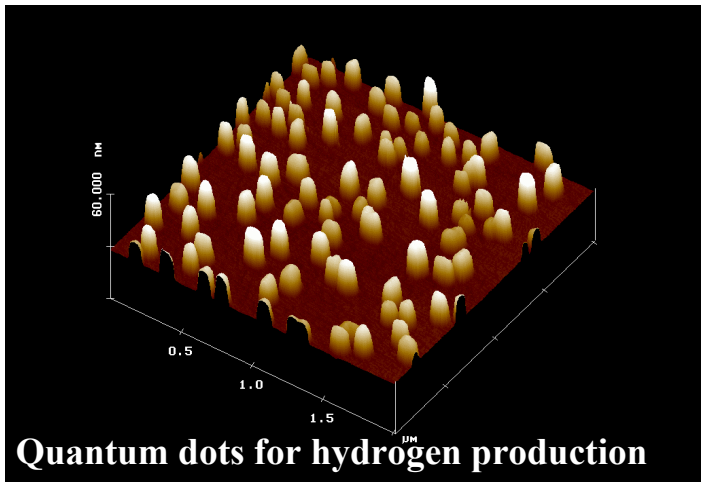
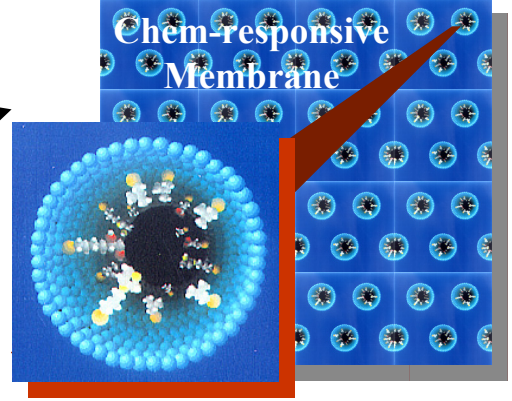
Energy Science and Technology Directorate

Micro Nano Breakthrough Conference
Portland, July 29th 2004

NSTI is a science initiative: Control of nanostructure is a gateway



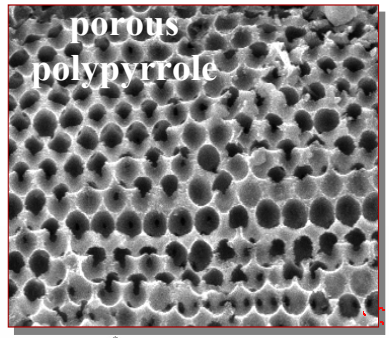




Tailored Response

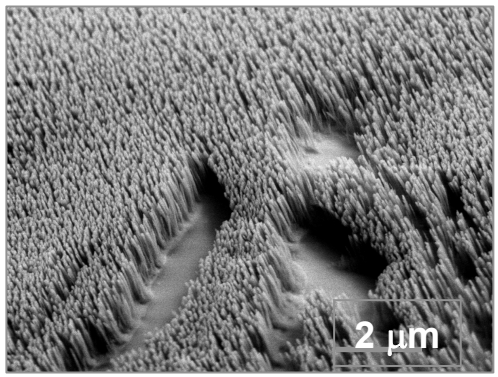
Functionalization

Ordered Porosity



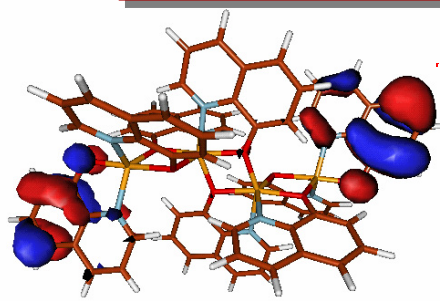
Self-Assembly

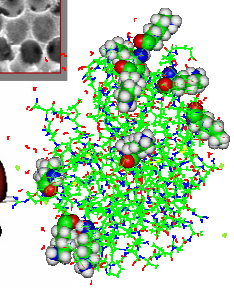
Clusters, Layers



MgO nanowires

Supra-Molecules

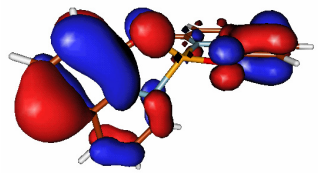




Enzyme

Synthesis

Molecules



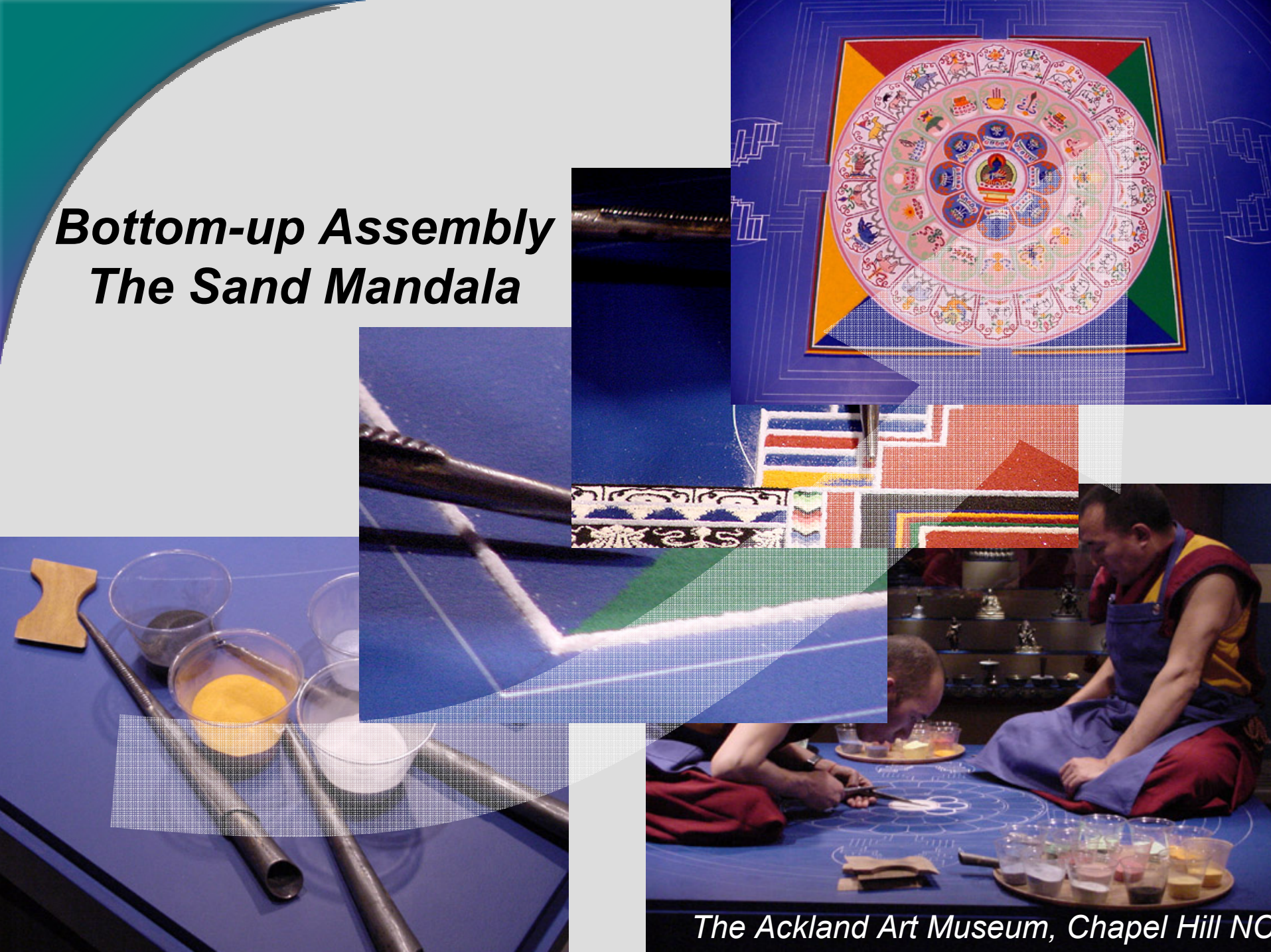
Orbital structure of Znq monomer and tetramer: enhanced electron transport in OLEDs

Atoms

Hierarchical Architecture

Degree of Functionality (Smartness of the Material)

Bottom-up Assembly
The Sand Mandala



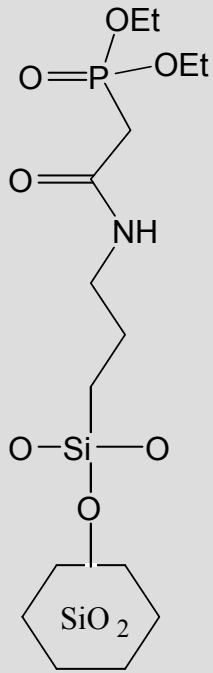
The Ackland Art Museum, Chapel Hill NC

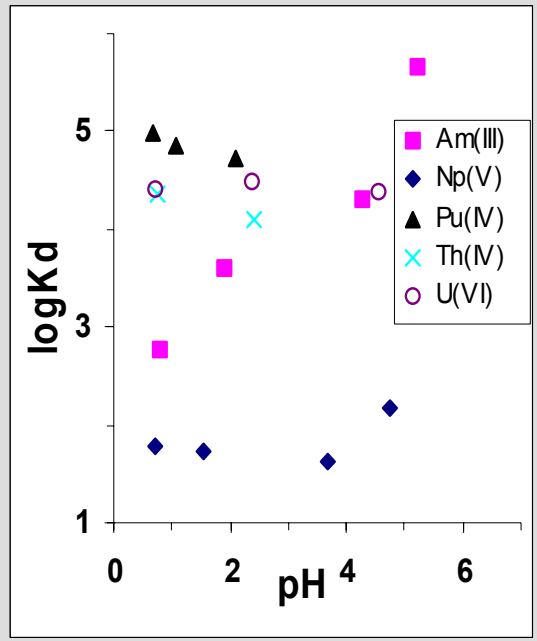
2 Monks, 2 weeks

Nanotechnology needs heterogeneous integration

Radionuclide Preconcentration and Measurement with SAMMS (Addleman)

- Design sorbent with highest possible selectivity AND affinity
  - Preconcentration 10^4- 10^7
- Integrate with material to support rapid sampling and assay





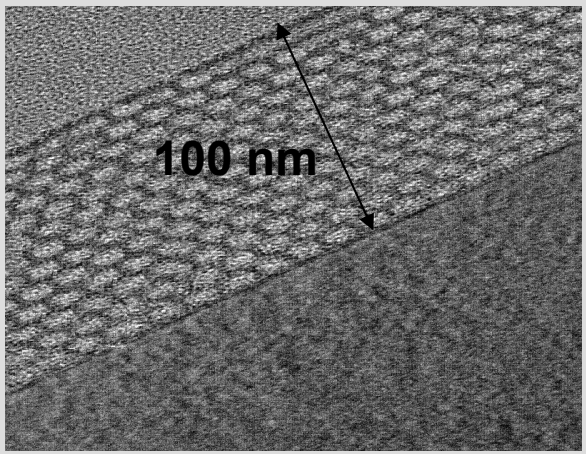
Ac-Phos* SAMMS

* acetamide diethylphosphonate moiety with 3-aminopropylsilane linkage

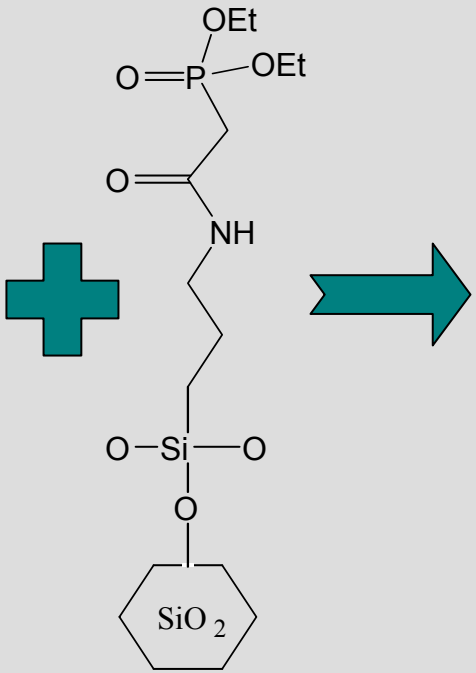
Chemical Communications, **2002**, 1374-1375.

Mesoporous Thin Films

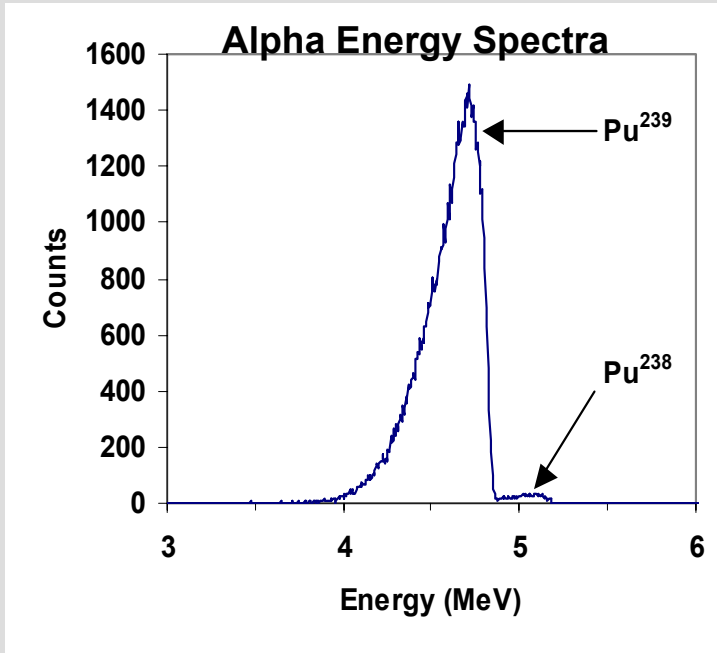
- Water stable, high surface area, chemically selective
- Increases surface area 50-50,000x
- Spin coating and thermal cure for uniform large area planar deposition
- IR Flash cure for irregular or thermally sensitive substrates



0.1- 3 μm thick, 1-10 nm pores, 600-900 m^2/g

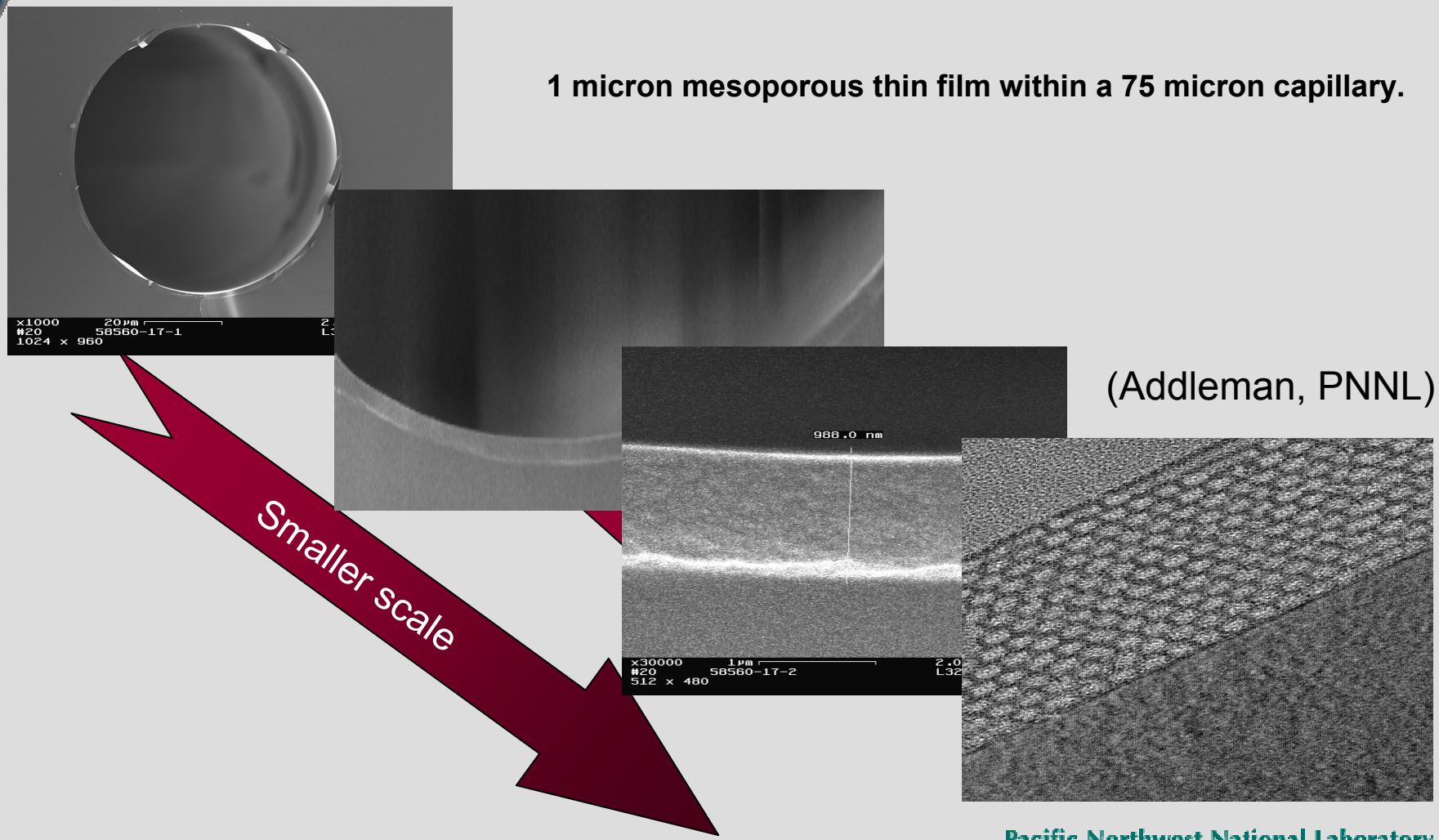


acetamide diethylphosphonate moiety with 3-aminopropylsilane linkage



1.8 μm thick, 8 nm pores, 620 m^2/gm,

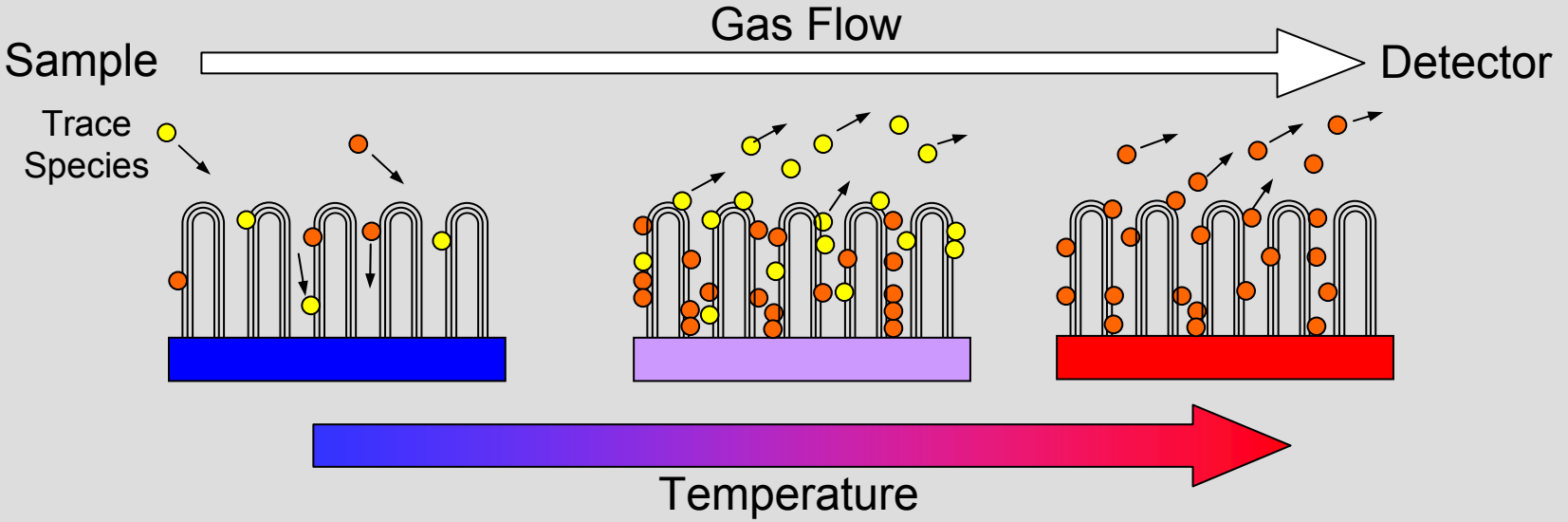
Hierarchical Structures
Thin Films in Capillaries



1 micron mesoporous thin film within a 75 micron capillary.

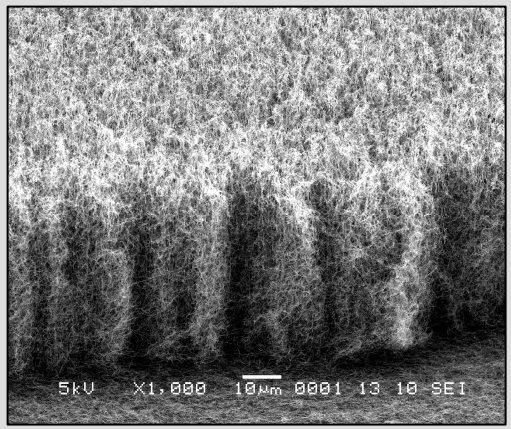
(Addleman, PNNL)

Preconcentration of Trace Chemical Signatures Using Carbon Nanotube Composites (Aardahl - PNNL)



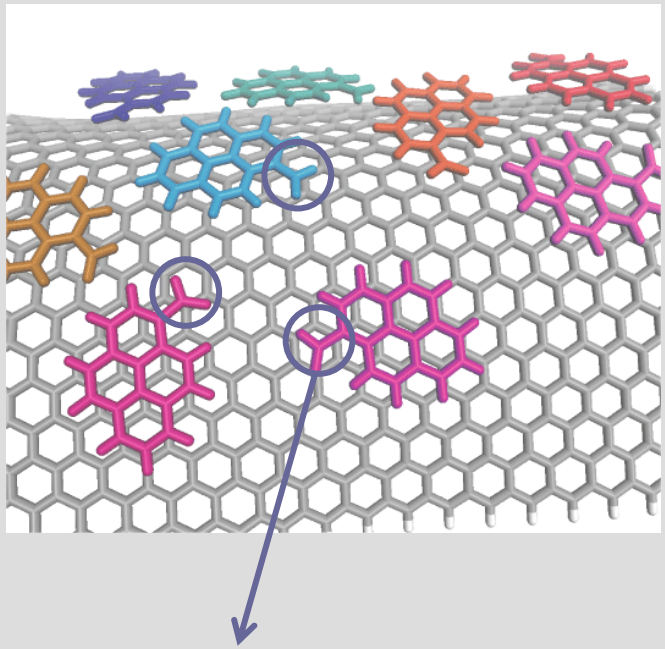
Why use CNTs?

- **High surface area** → compact → dead volume reduction, temperature uniformity, better separation resolution
- **High thermal conductivity** → fast cycling and reduced detection intervals

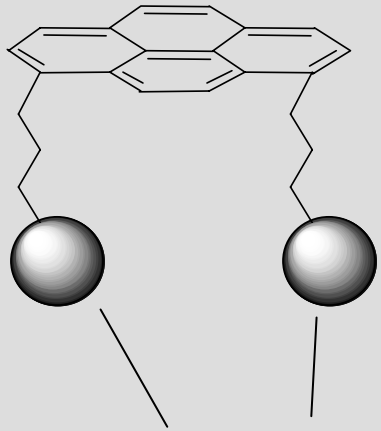


Non-Covalent Functionalization

Anchor-based functionalization
(Chen *et al.* (2001) *JACS* **123** 3838



Selective groups for O=P-R binding

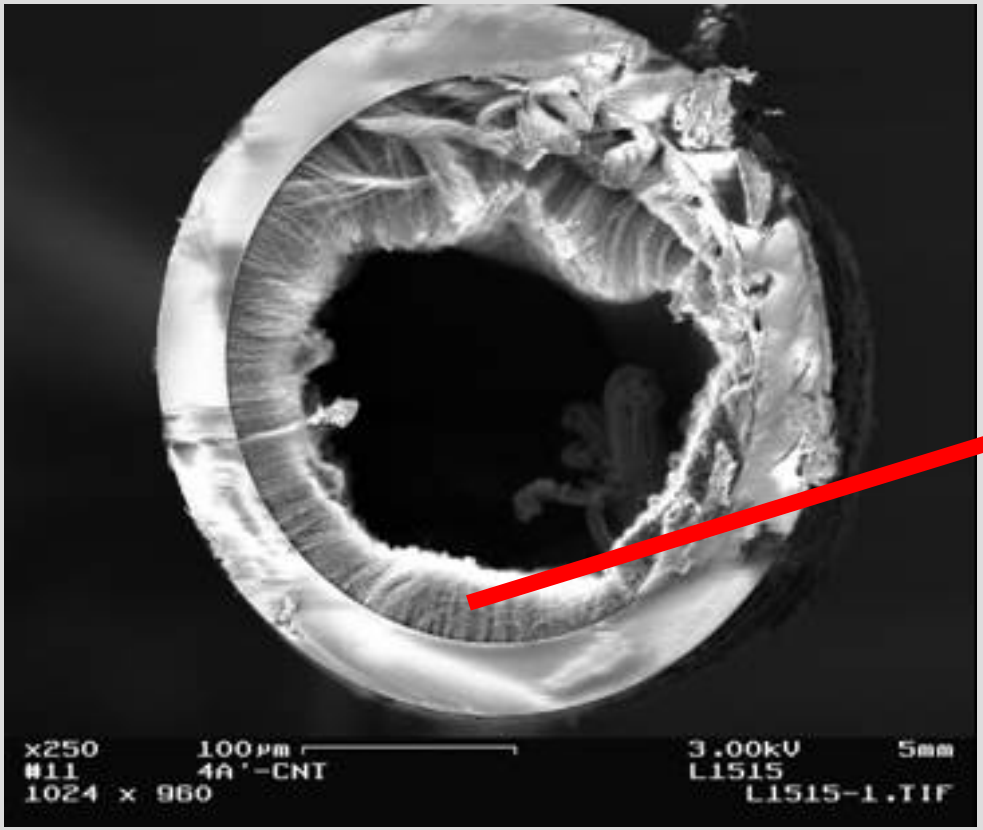


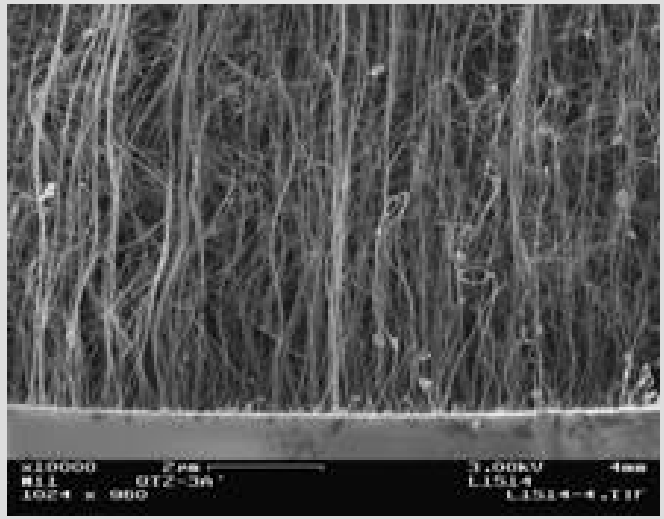
Selective groups tethered
to molecular anchor

Synthesis in Progress
Unclear how many functional groups
are possible for a given pyrene

Joint Institute for Nanoscience:
Collaboration with Prof. L.R. Dalton

Carbon Nanotubes in Capillaries via CVD

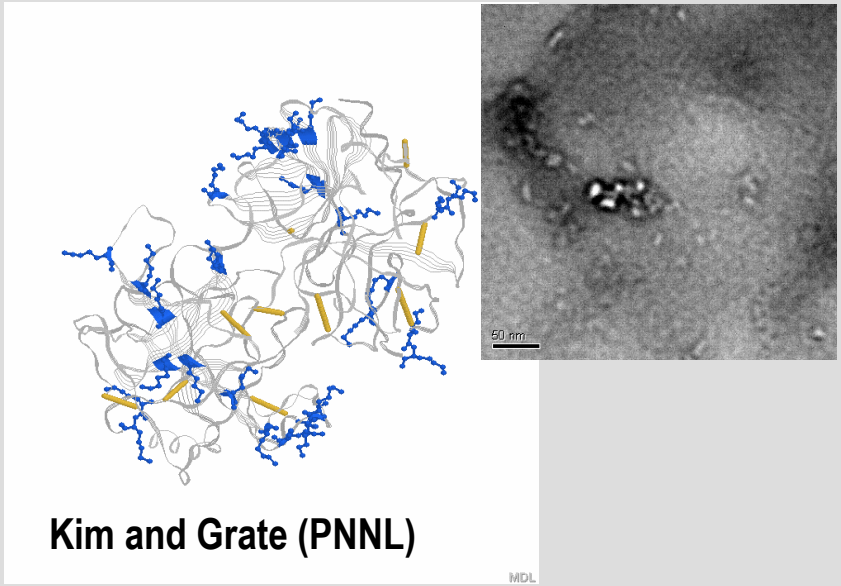




Quartz capillary coated with carbon nanotubes

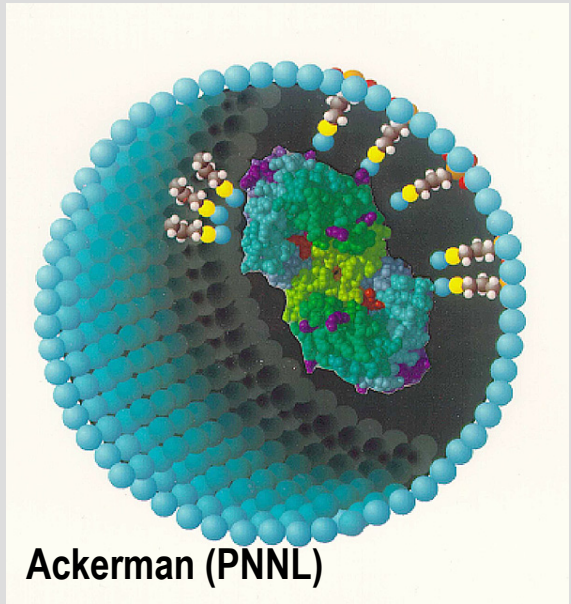
Nanobiology at PNNL: Enzyme based machines

Enzyme Confinement: Significant Activity and Stability Enhancements



Kim and Grate (PNNL)

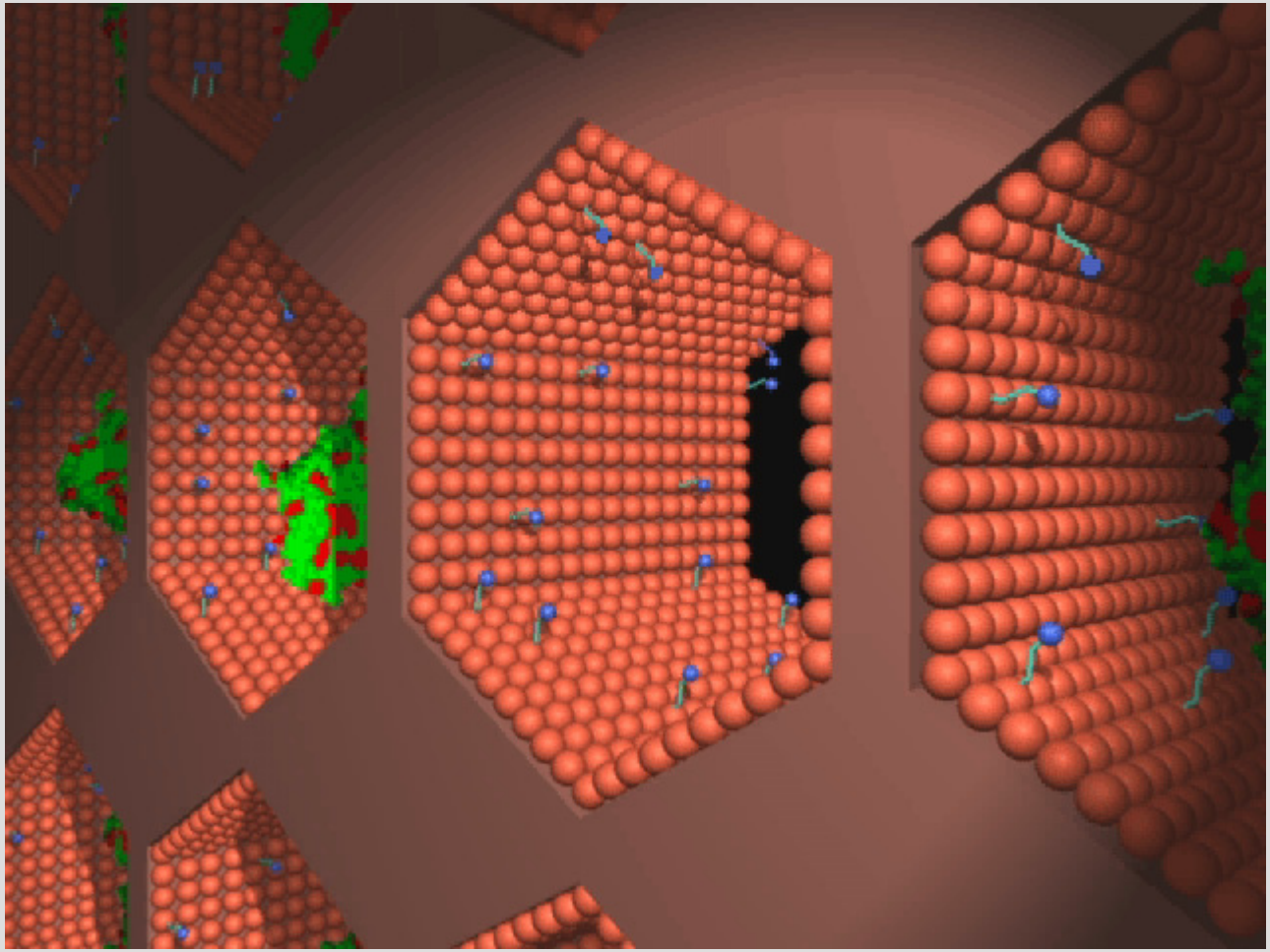
Single-enzyme nanoparticles “armored” with a porous composite organic/inorganic network of less than a few nanometers thickness



Ackerman (PNNL)

Single enzymes embedded in mesoporous silica and tethered to surfaces.

Enzyme Stabilization in Mesopores

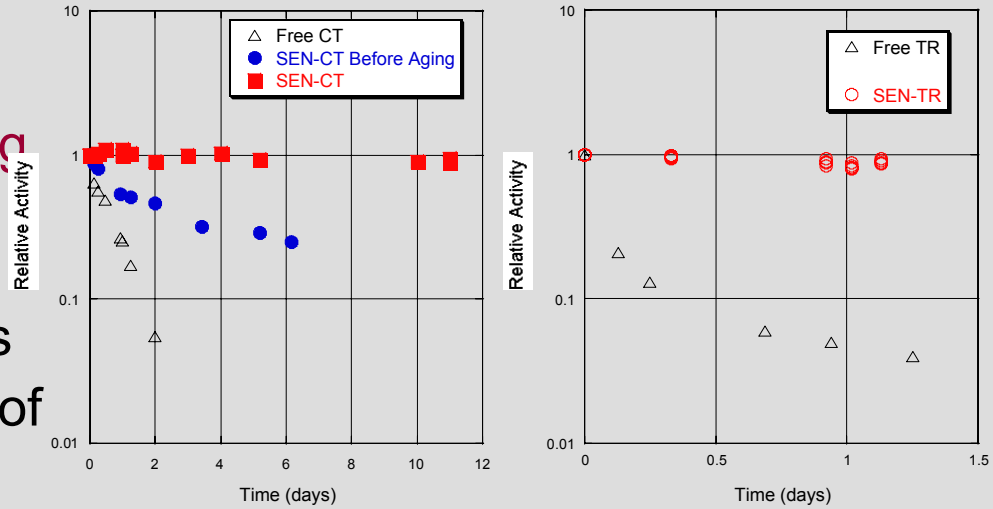


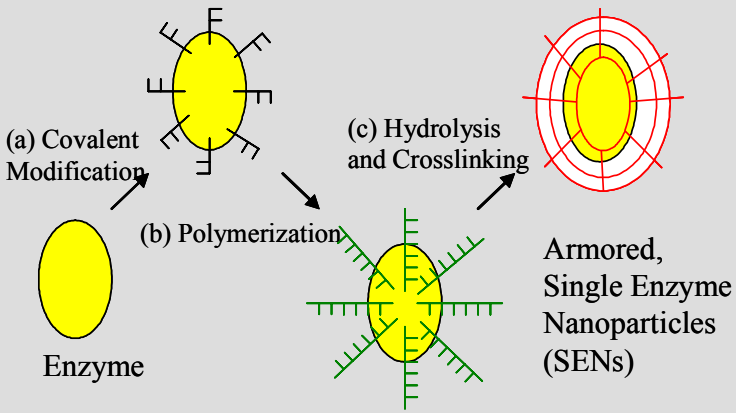
e.g. organophosphorous hydrolase

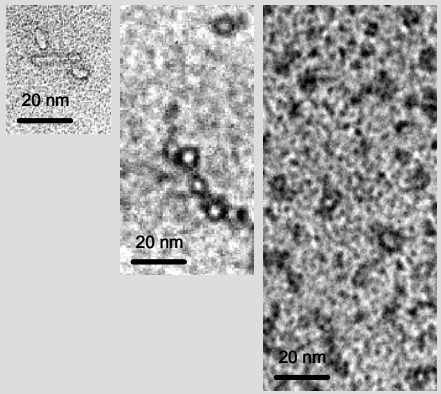
Eric Ackerman (PNNL)

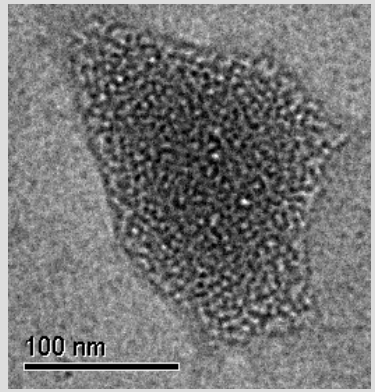
Armored Enzymes

- Stabilization of two proteases (CT and TR)
- Controlled thickness of coating
- No mass transfer limitation through the armor shell
- Proteolysis of various proteins
- Resistance to the proteolysis of unarmored proteases



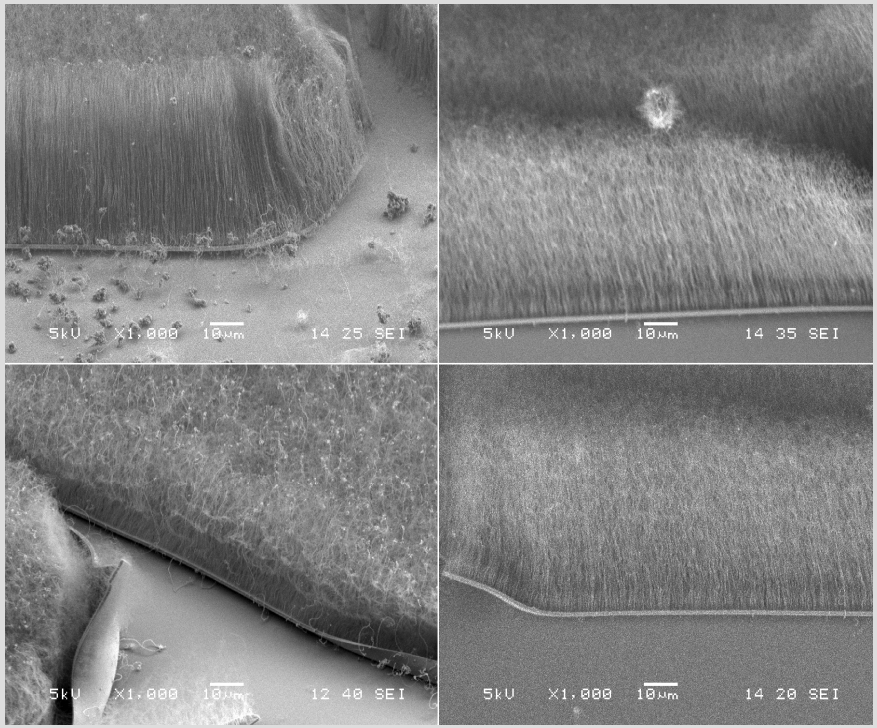






Tyrosinase on 3-D CNTs (Carbon Nanotubes)

- Initial Activity of Tyrosinase on 3-D CNTs



	Linker	Tyrosinase Activity	Ratio to Quartz
No CNTs (Quartz)	APTES-GA	1.7	1
CNT	EDC	25	15
CNTs	Pyrene	55	32

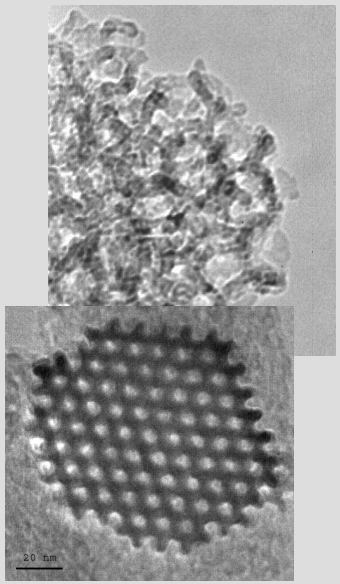
SENs in Nanostructured Matrices

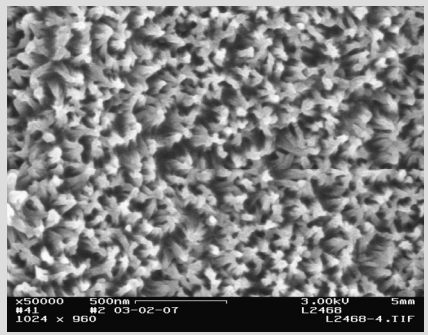
Mesoporous Silica (MPS)

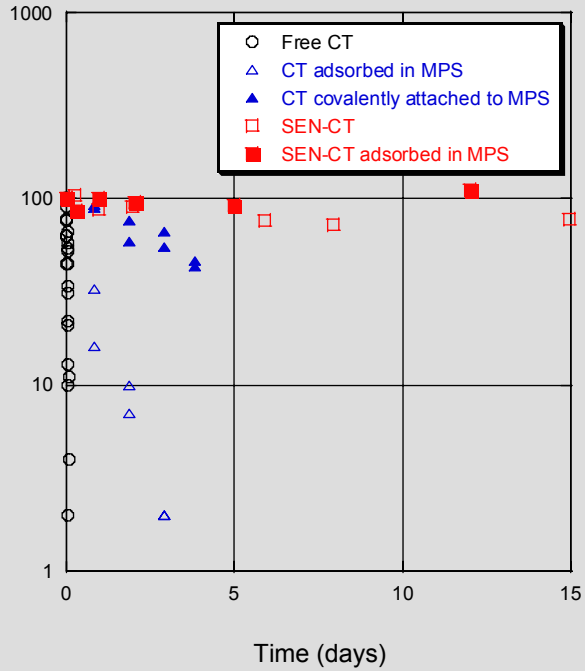
- SEN-CT adsorbed in MPS was more stable than free SEN-CT

Conductive Polymer Nanowires

- Polyaniline nanowires
- Mechanistic studies on nucleation and growth

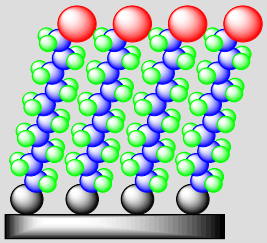




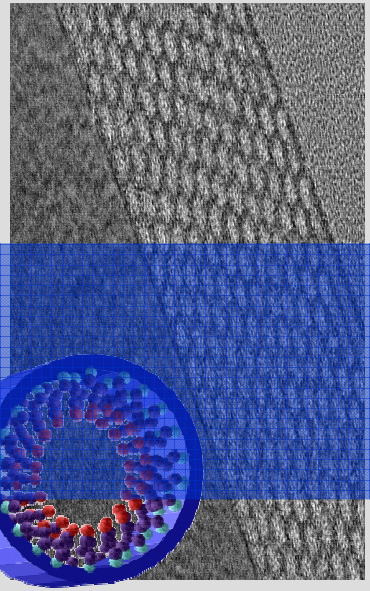


“Deca-scale” Integration (10^{10})

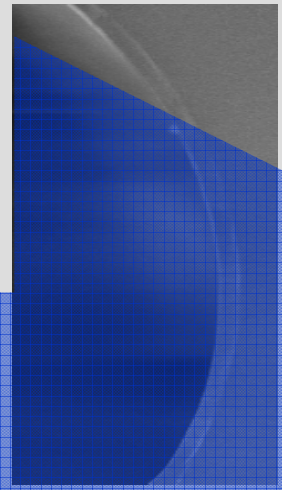
Active groups selective for target material.

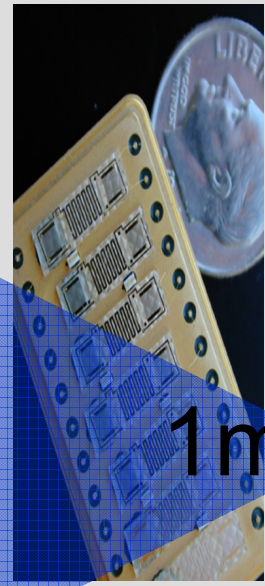


Mesoporous supports



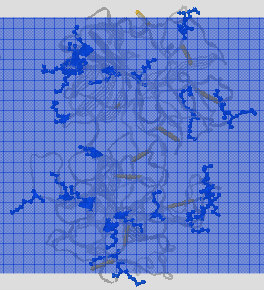
Capillary systems



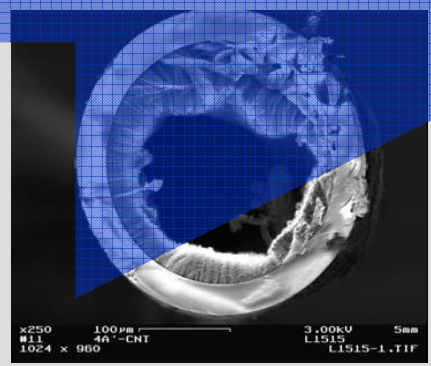


Enzymes

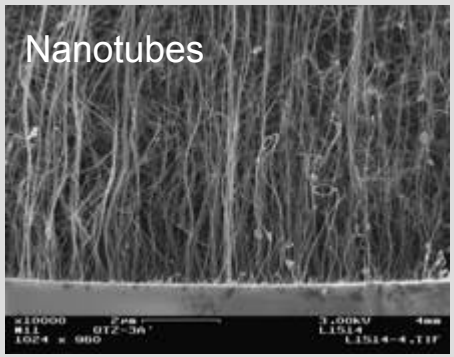
1Å



1m

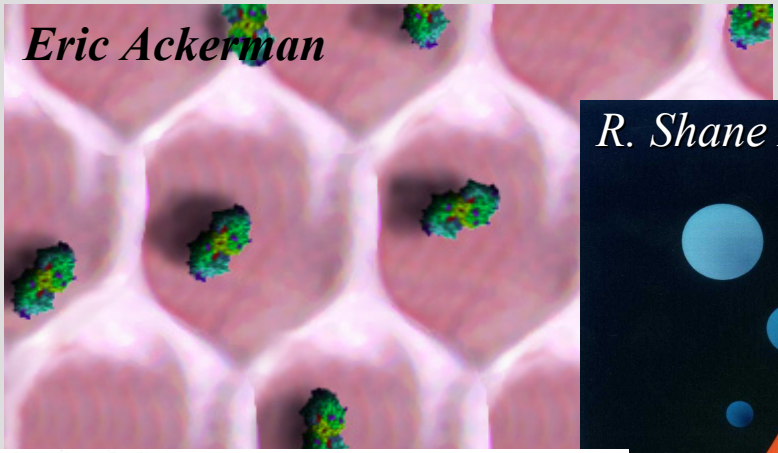


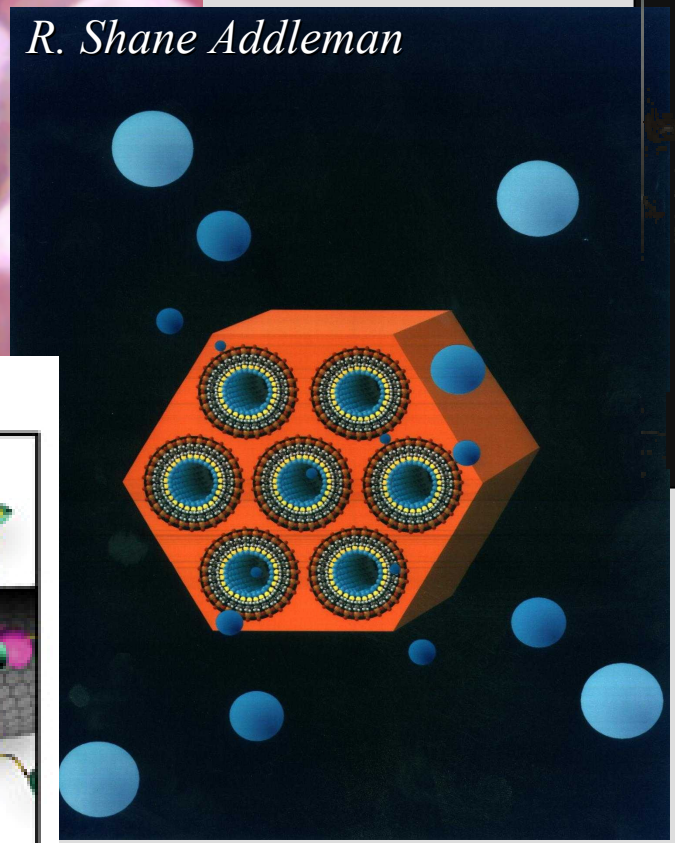
Nanotubes

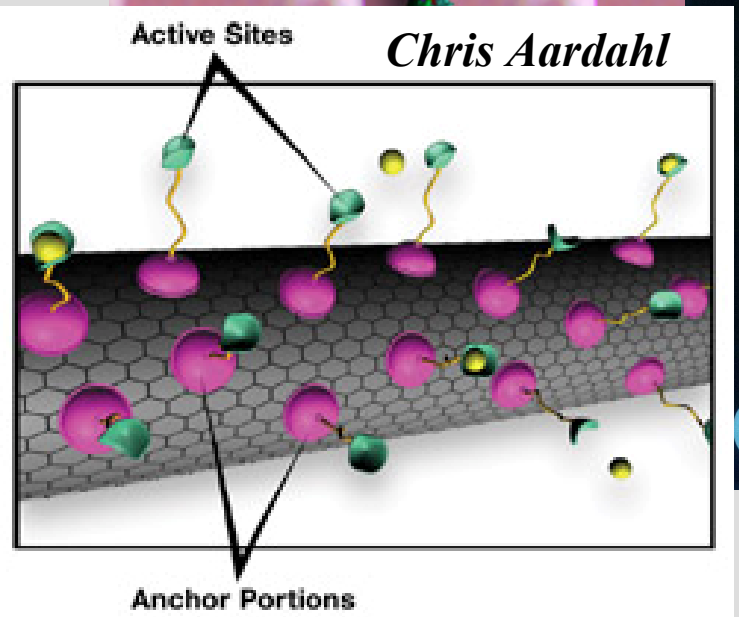


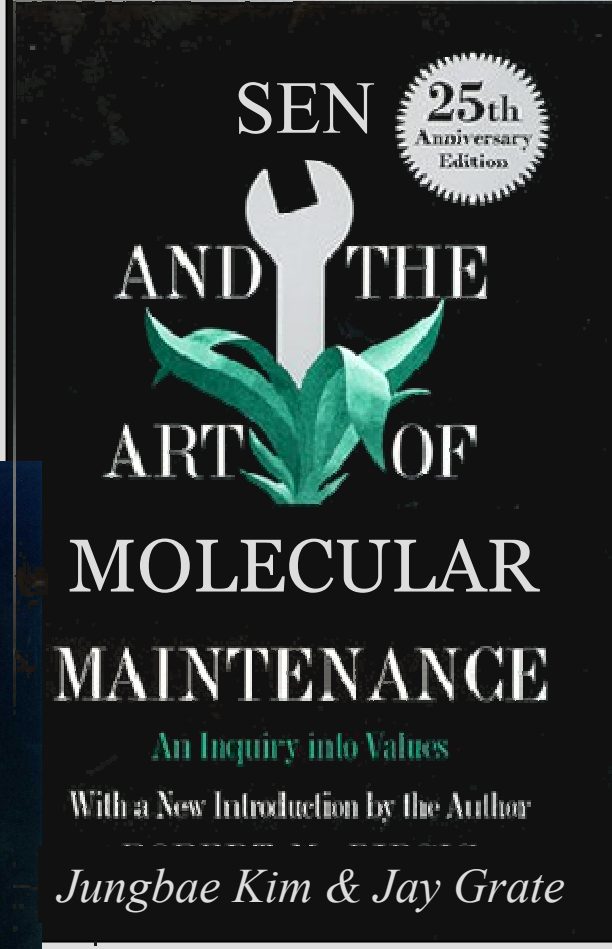
Enhanced Sensor Arrays

Thanks to the cast of dozens…









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