

# TOWARDS SUSTAINABLE MEMBRANE MATERIALS: GO-HAL!!!

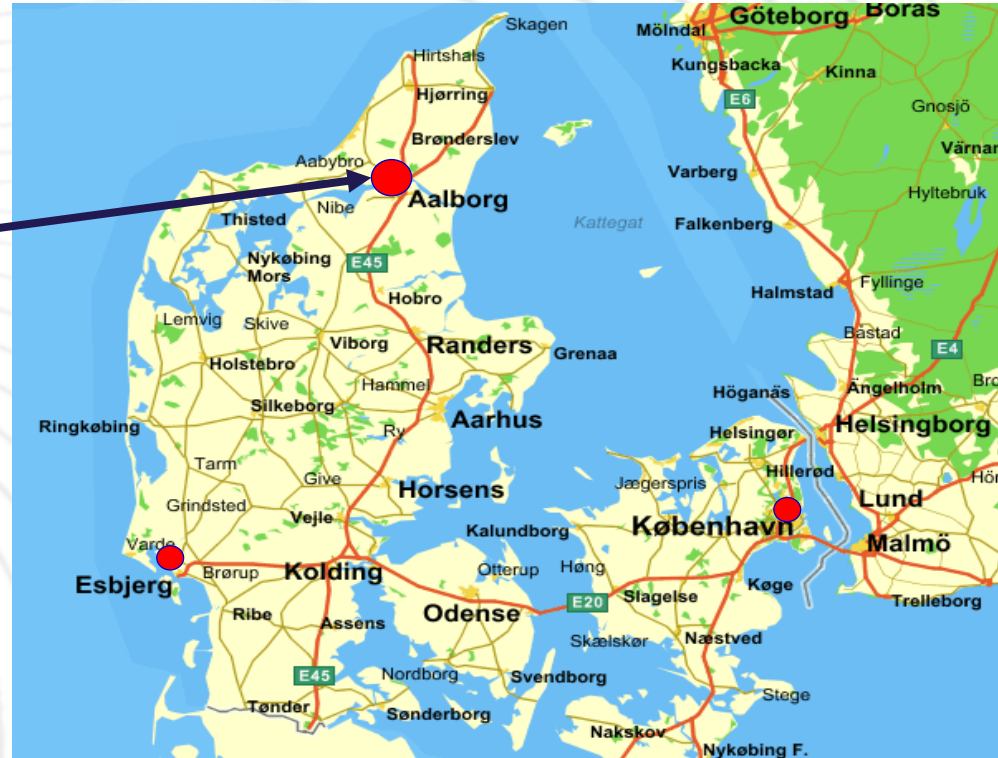
VITTORIO BOFFA

[vb@bio.aau.dk](mailto:vb@bio.aau.dk)



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# Aalborg University – Department of Chemistry and Bioscience

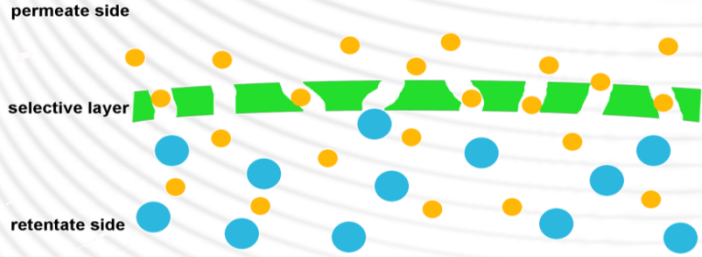


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Aalborg Centre for Problem Based Learning  
in Engineering Science and Sustainability  
under the auspices of UNESCO

# Membrane filtration

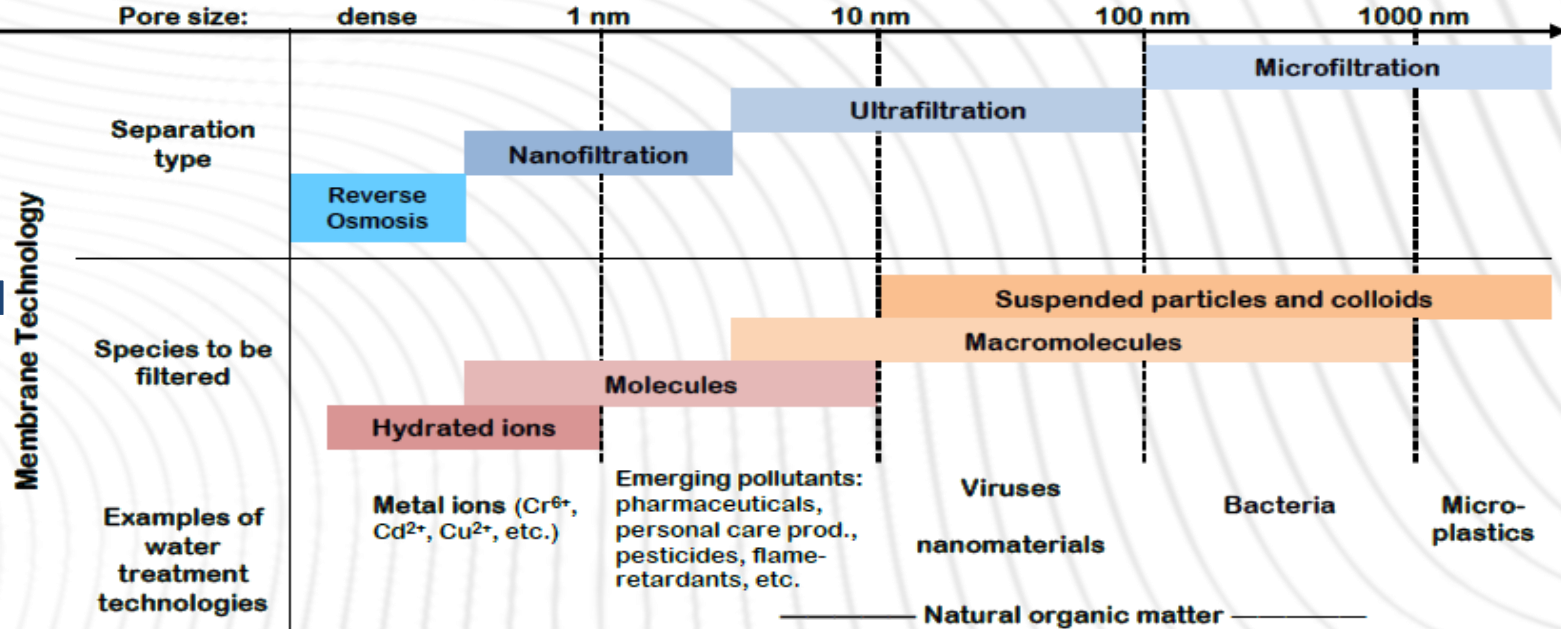


**Membranes are selective barriers**

They allow removing pollutants

- with low energy consumption;
- no additional chemicals are needed!

Trend towards functional membranes ←



# Inorganic membranes vs polymeric membranes

## Inorganic membranes

### PRO

- Inherent hydrophilicity:
  - high water flux
  - low fouling
- Resistant to high temperature and chemicals:
  - easy to clean and to sterilize
  - long operational time (> 15 years, 2-3 times longer than polymeric membranes)

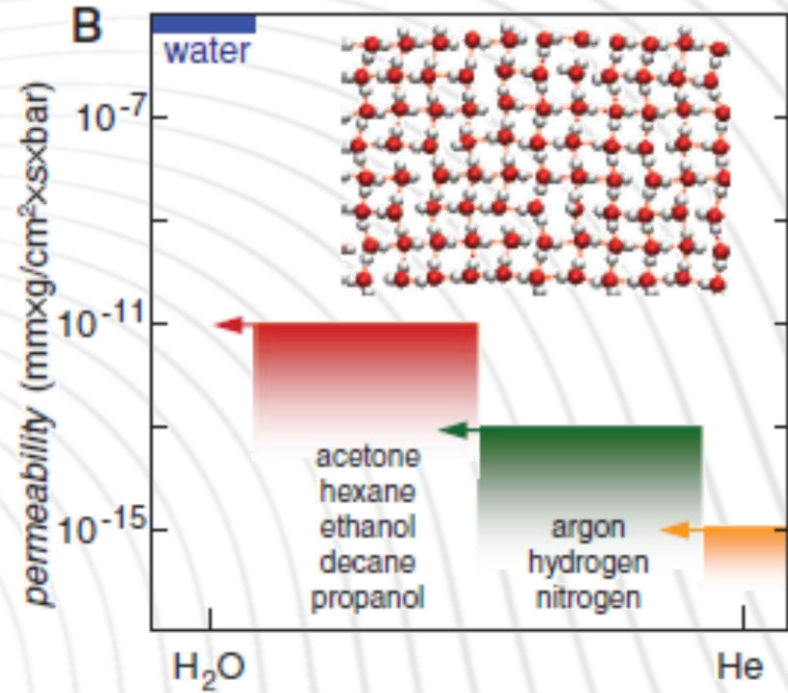
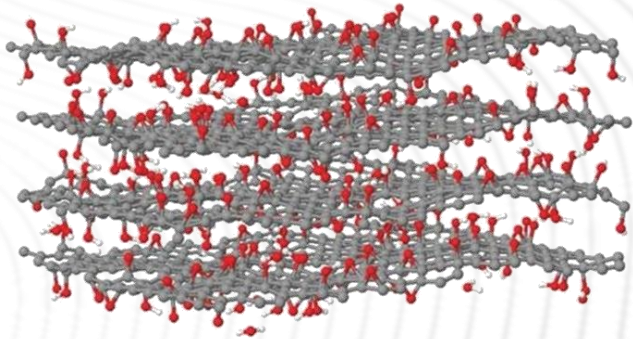
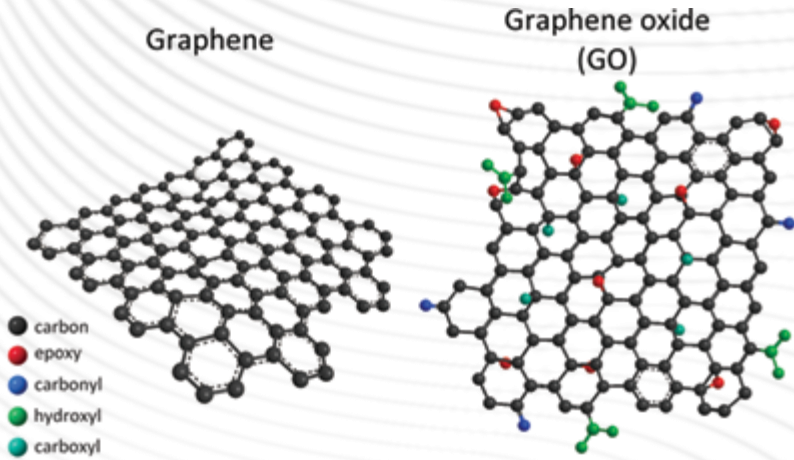


Trend towards increasingly environmentally, economically, and socially sustainable materials

### CONTRA

- Material cost + energy consumption for fabrication > 3 times higher than polymeric membranes
- Low filtration area densities.

# Graphene oxide membranes



R.R. Nair et. al. *Science* 335 (2012) 442.

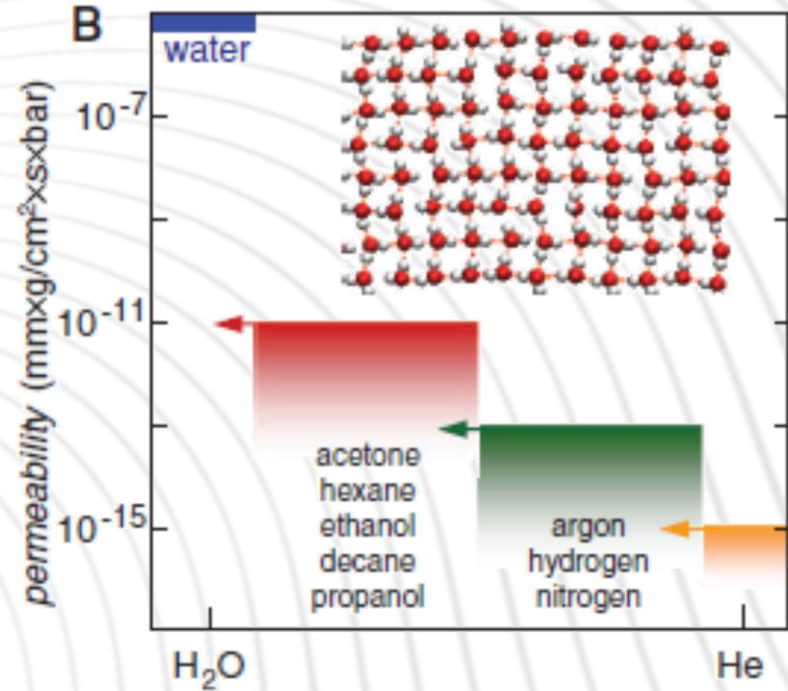
# Graphene oxide (GO) membranes

## Advantages:

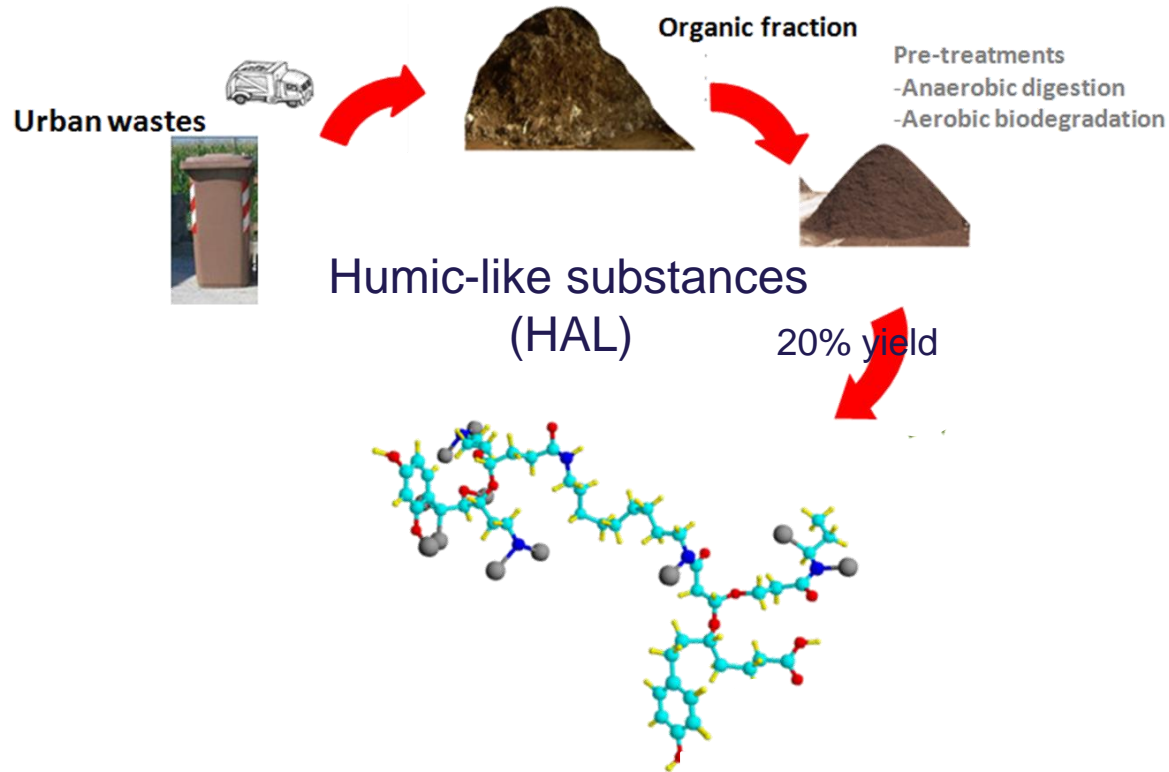
- Outstanding water permeability
- High water selectivity
- Easy to process

## However,

- Low stability under cross-flow filtration
- Thermal densification



R.R. Nair et. al. *Science* 335 (2012) 442.



## Valorization of the organic urban refuse

# GO & HAL

Chemical similarity:  
carbon backbone functionalized  
with oxygen-containing moieties

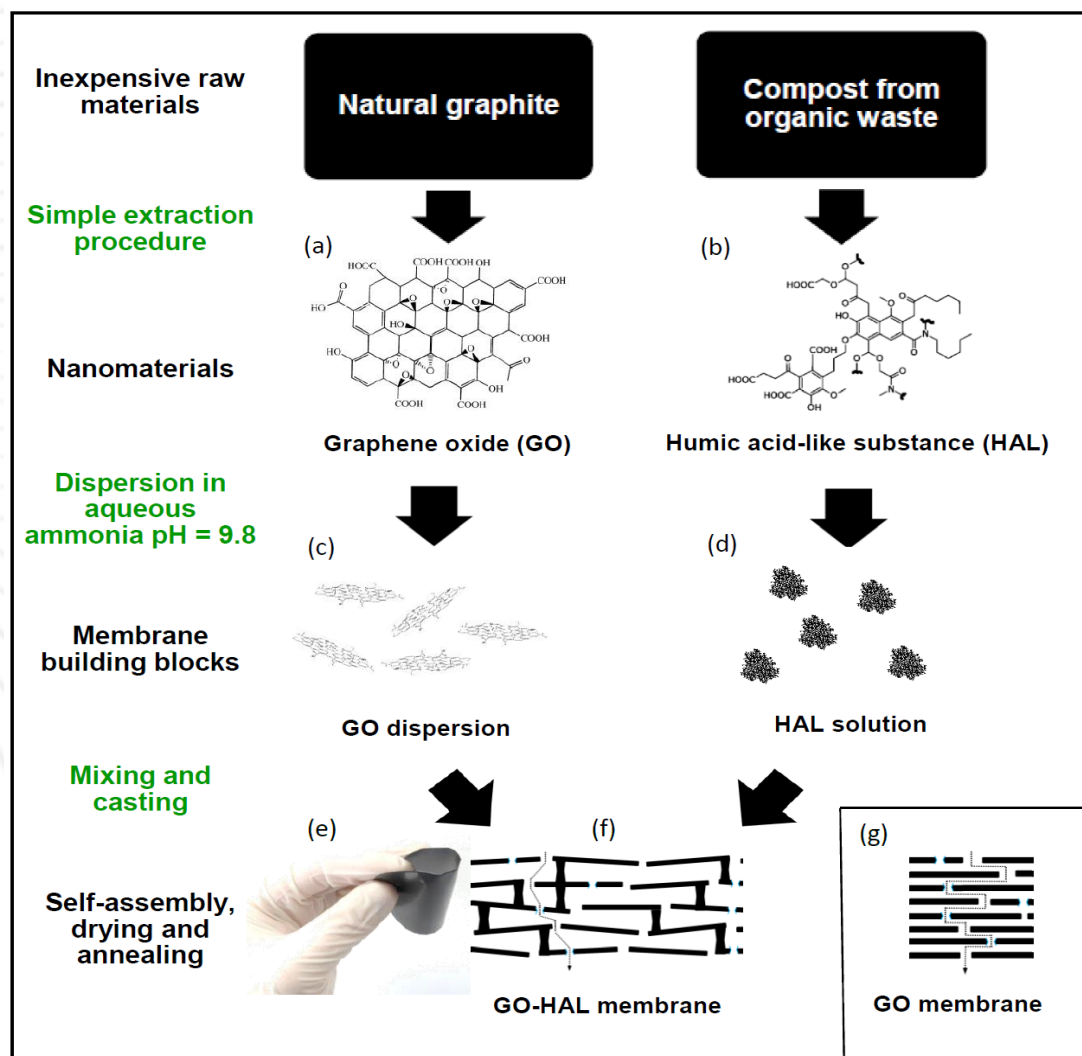
## Structural differences:

GO → 2D layers

HAL → 3D branched structure

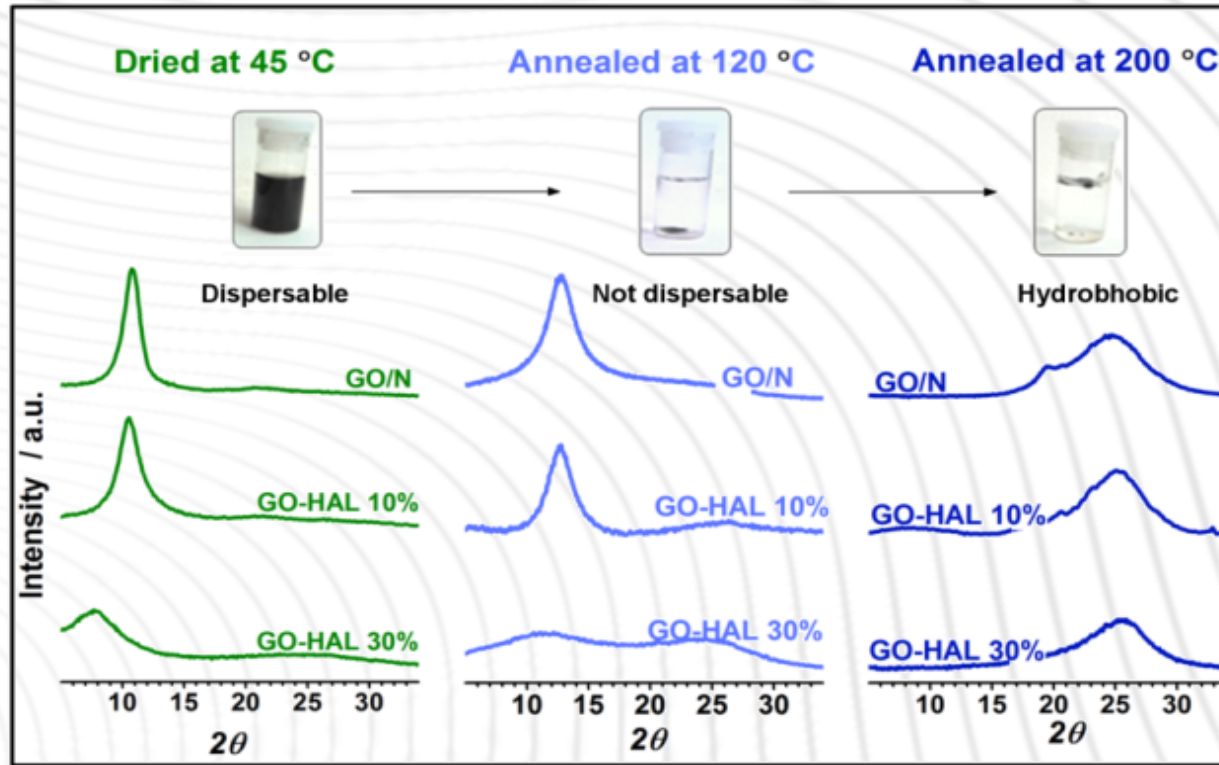
## Hypotheses:

- HAL can intercalate GO layers, thus inducing structural disorder
- HAL can increase water permeability of GO membranes upon thermal stabilization

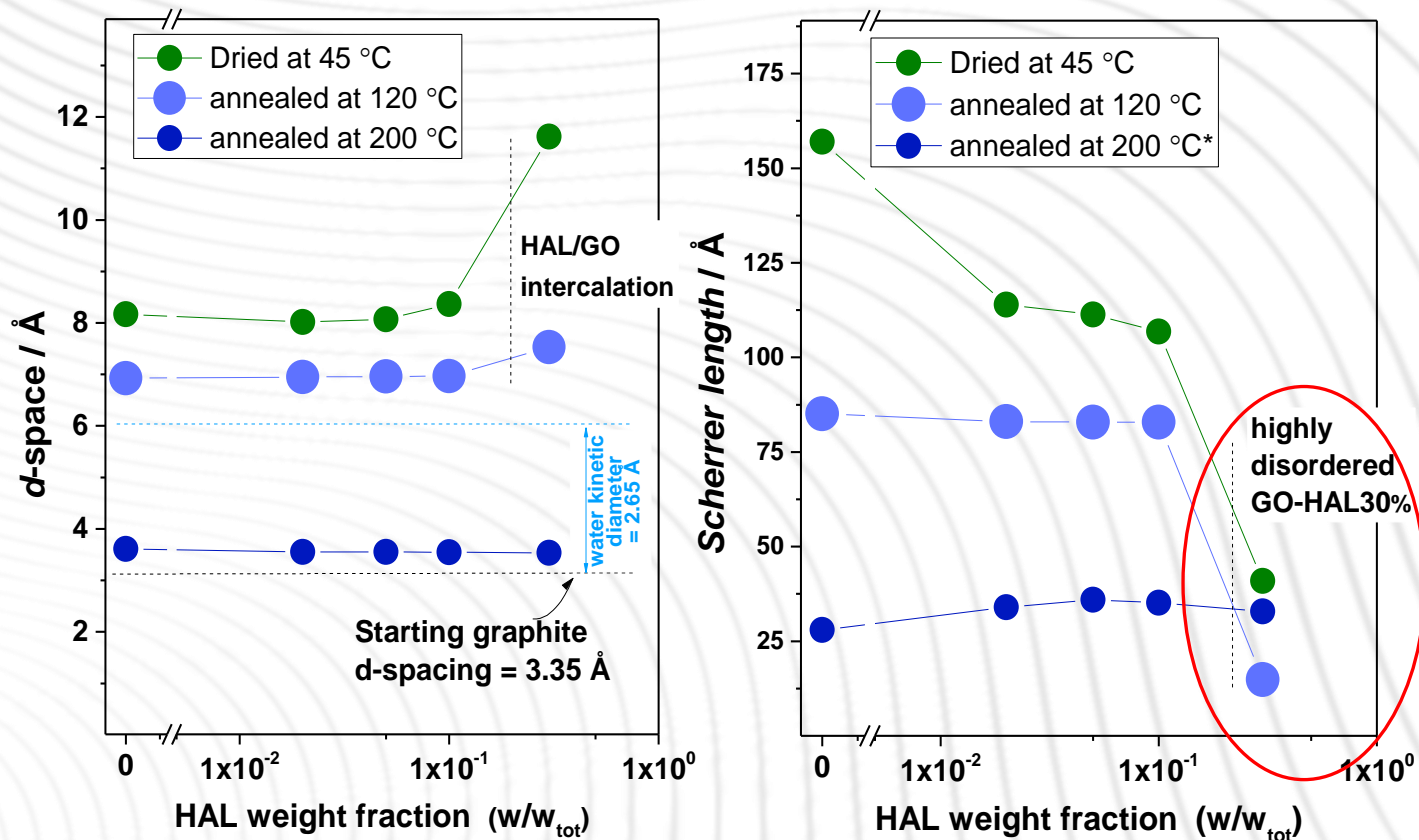




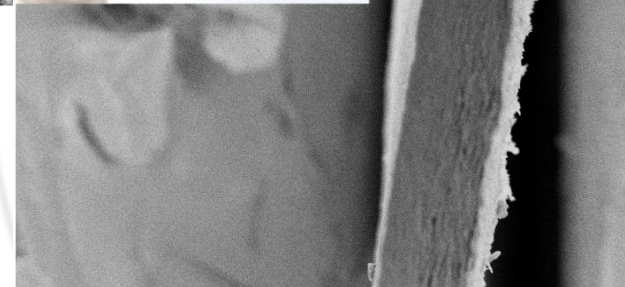
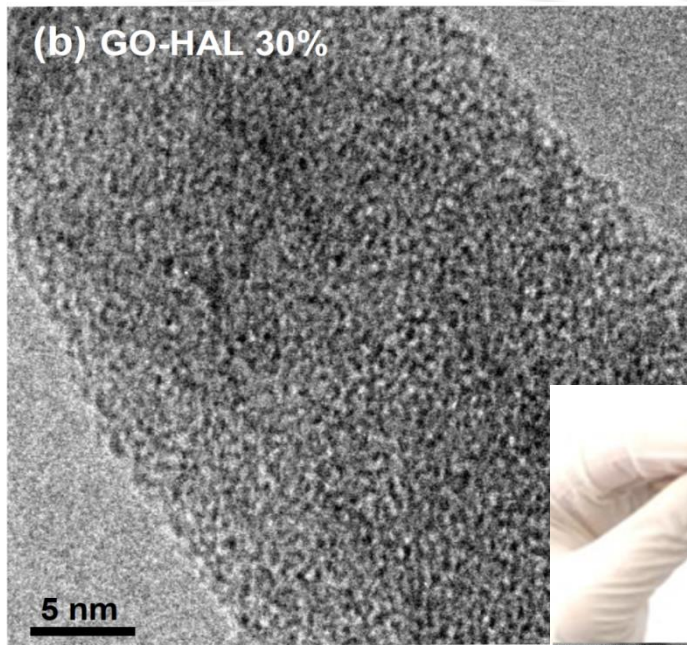
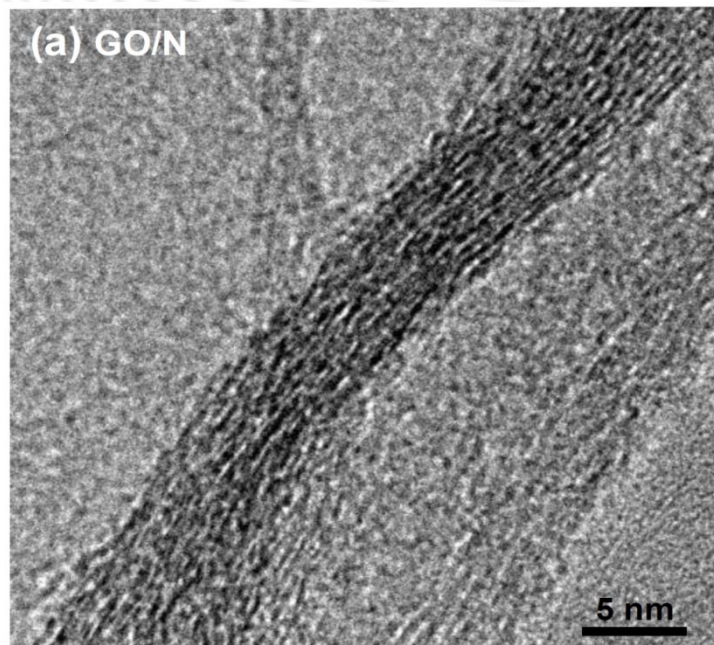
# order vs disorder



# order vs disorder



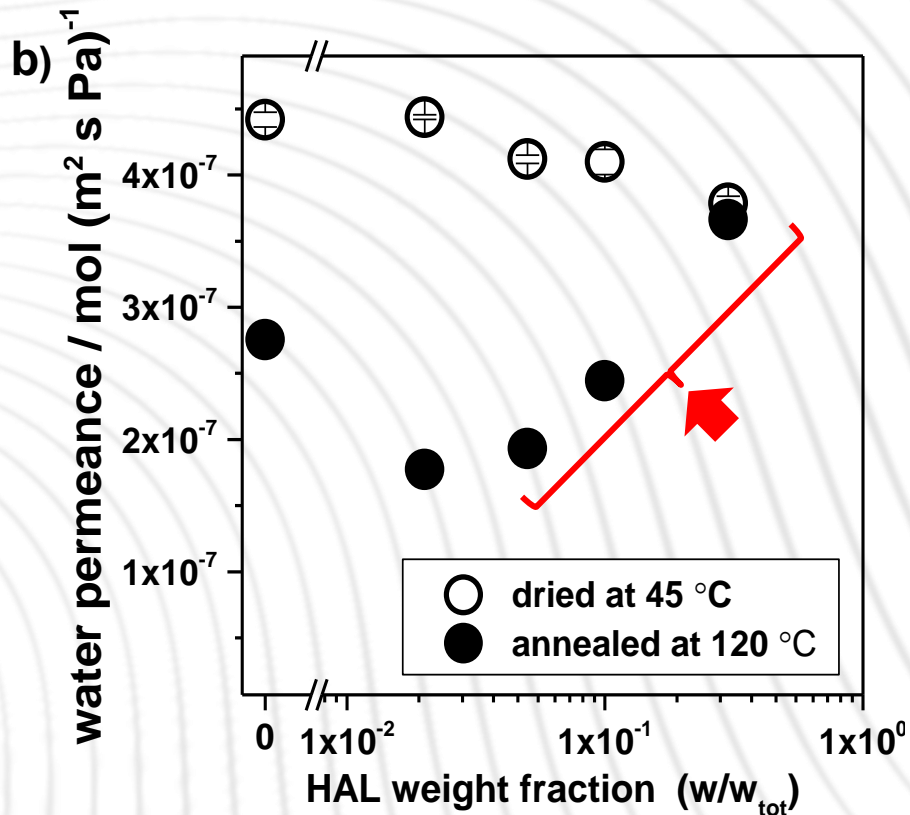
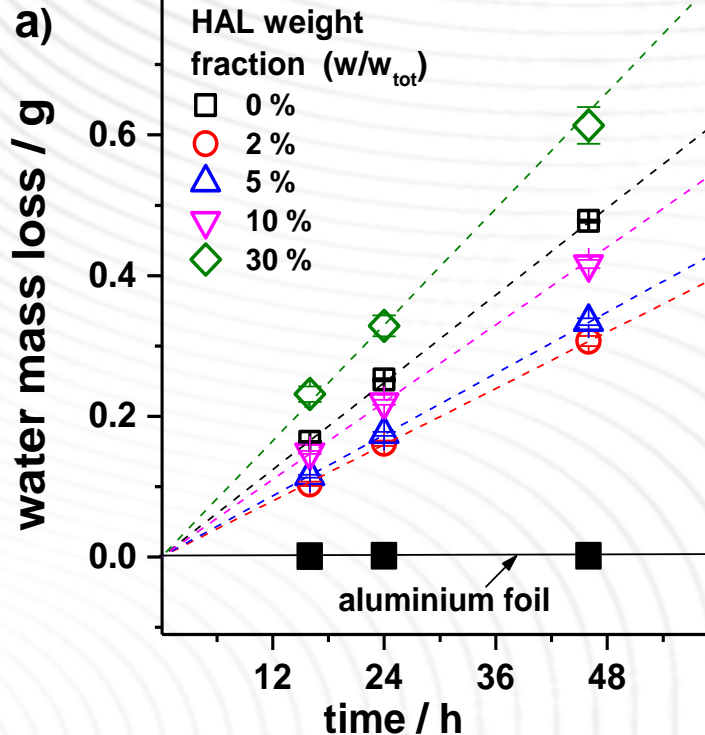
# order vs disorder (after annealing at 120 °C)



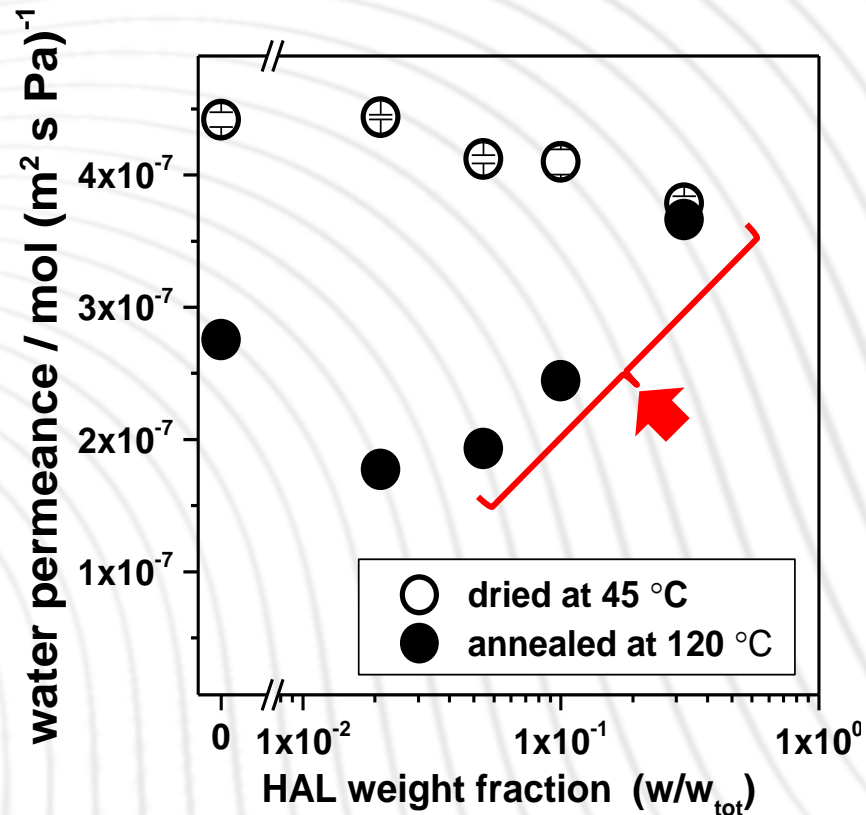
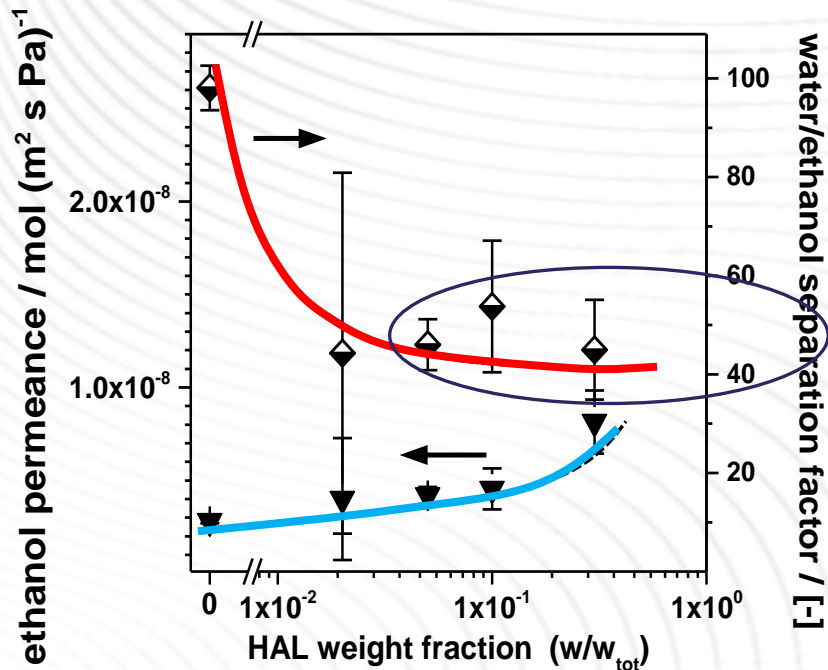
V. Boffa et. al. *Carbon* 118 (2017) 458

# Membrane water permeability

Annealed at 120 °C



# Membrane Perm-selectivity



## ***In summary***

- **GO-HAL films were fabricated via a facile process**
- **After annealing at 120 °C:**
  - **GO-HAL are stable in water (dispersion tests)**
  - **GO-HAL30% has water permeance 33% higher than pristine GO**
  - **GO-HAL30% has an ideal water/ethanol selectivity of 45**
- **GO-HAL appear to be highly promising for alcohol dehydration technologies**
- **Disorder in the GO-structure is a crucial parameter for GO membranes**

# Is GO-HAL a sustainable membrane material?

## ***Yes, because:***

- **Cheap, natural, and abundant starting materials: graphite and organic compost**
- **Recycling organic urban waste**
- **Easily, scalable and water-based synthesis and processing**
- **Heat recovered at the membrane end-of-life**

## ***But, there are some challenges:***

- **Hummers' GO can contain 1-5 w% sulfur**
- **New green methods for GO production**
- **Valorization of the insoluble residue of HAL extraction**

# Acknowledgments



- P.E. Mallon (University of Stellenbosch, South Africa)
- G. Magnacca (Turin University, Italy)





# HUMIC SUBSTANCES

## Vital resource affecting food chains

- directly
- indirectly (turbidity, pH, metal chelation,...)

## Technological properties

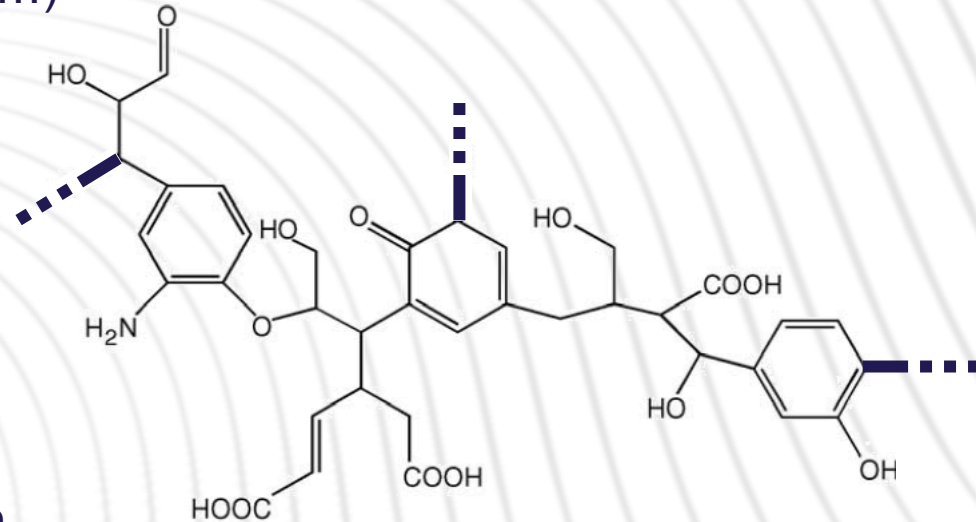
- polyelectrolyte
- supramolecular structures, micelles

## Availability?

Humic carbon in the oceans is comparable to the amount of CO<sub>2</sub> carbon in the atmosphere.

**But...** diluted (typically mg/L)

## Virtual fragment of aquatic humic acid



adapted from

S. McDonald et al., *Analytica Chimica Acta* 527 (2004) 105