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Molecular imprinting-based chemical sensors for the detection of monocyclic aromatic compounds

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Introduction

The development of biologically inspired recognition materials, capable of selective target binding/rebinding with comparable efficiency to substrate-enzyme or antibody-antigen interactions is a key point in the fabrication of chemical sensors.

Molecularly imprinted polymers (MIP) are a new class of synthetic receptors that can be used as robust and cheap sensing elements. When associated to an appropriate transducer, they can lead to very efficient and easy-to-use chemical sensors.

The Principle of Molecular Imprinting

Molecular Imprinting exploits the simple, but elegant, principle of using elements of a target molecule to create its own recognition site. This is achieved allowing a template, e.g. an analyte of interest or an analyte closely related to it, to form complexes in solution with one or more polymerisable receptors that have the possibility to interact with this template through hydrogen bonds, ionic interactions... Then, a highly cross-linked polymeric matrix is formed around the template-functional monomer complexes by adding an excess of cross-linking monomer, thus "freezing" these interactions in such an arrangement that would allow subsequent rebinding of any analyte that has the correct size, shape and functionality to match the requirements of the so-called *binding site* (Figure 1)

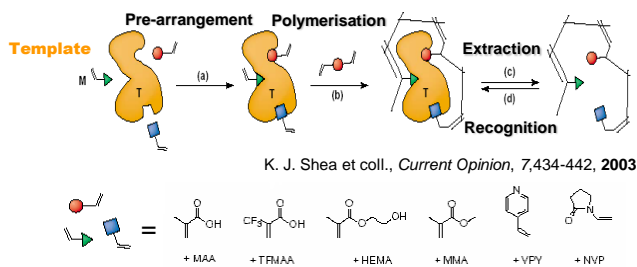
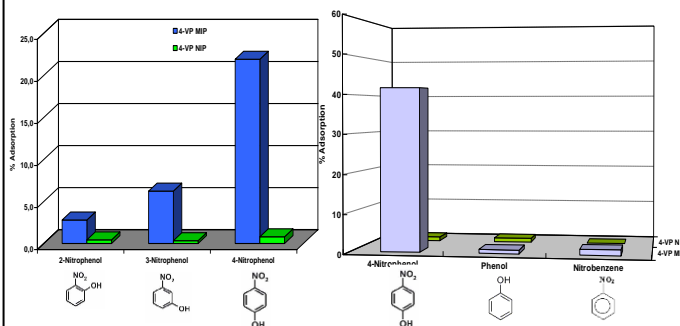


Figure 1. Schematic representation of non-covalent imprinting process

- (a) Pre-polymerization complex formation between template (T) and functional monomers (M).
 (b) Co-polymerization with an excess of crosslinking monomer.
 (c) Extraction of template from imprinted binding site.

MIP screening for 4-NP & interferences

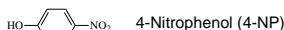


Procedure:

10 mg of polymer were incubated in the presence of three different analytes ($[Analyte]_0 = 1 \text{ mM}$). After 24 hours, the supernatants were analyzed and quantified by gas chromatography. The pourcentages of adsorbed analyte are obtained by subtraction.

The 4-NP imprinted polymer showed a markedly stronger affinity for the template 4-NP, separating 4-NP from the other nitrophenols isomers. A similar result was obtained when phenol or nitrobenzene were tested as interferences.

Structure of the template/target molecule



4-nitrophenol is included in the list of Environmental Protection Agency priority phenolic pollutants (<http://www.epa.gov>).

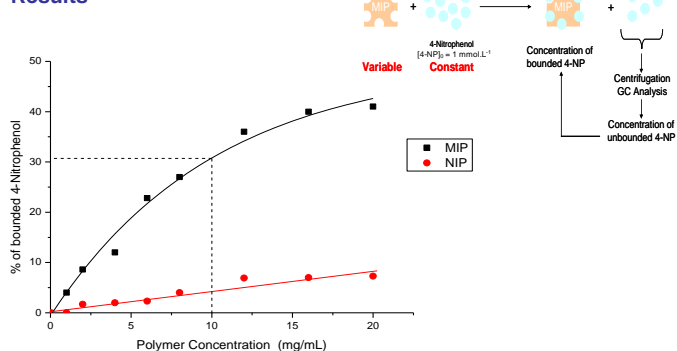
Method of preparation

MIPs were prepared by free radical polymerisation initiated thermally (AIBN at 60°C for 24h) in the presence of 4-vinylpyridine (4-VP) as functional monomer, ethyleneglycol dimethacrylate (EGDMA) as cross-linker and 4-NP as template in acetonitrile.

After polymerisation, monoliths are obtained. Once ground, samples require to be washed, dried and sieved before use.

A controlled polymer (NIP), prepared in the absence of the template, was also synthesised and tested according to the same procedures.

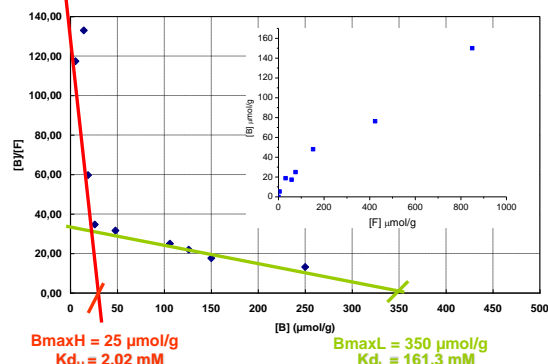
Results



Scatchard Plot

Scatchard Plot for 4-NP uptake by MIP-4VP was obtained from the binding curve (inset; the value above 200 $\mu\text{mol/g}$ are subject to high experimental error).

Two distinct linear regions with decreasingly steep slopes may be discerned: left most region, high affinity sites, 25 $\mu\text{mol/g}$ MIP, $K_{dH} = 2.02 \text{ mM}$; rightmost region (the least precise), 350 $\mu\text{mol/g}$ MIP, $K_{dL} = 161.3 \text{ mM}$.



Conclusions

An artificial receptor for 4-nitrophenol has been synthesised and tested in solution. A good selectivity for the target molecule has been shown despite of an heterogeneous distribution of binding sites.

Future work: - assess polymer performances in gas phase.
 - associate the synthetic receptor with a transducing device (acoustic and optical devices are currently under study)