

Werner Prize

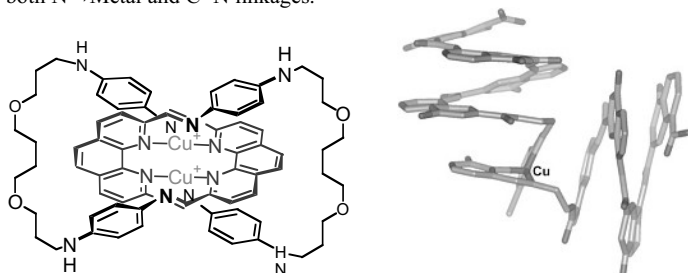
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From Simplicity to Complexity via Subcomponent Self-Assembly

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The creation of complex structures from simple building blocks *via* thermodynamic self-assembly requires an understanding of the rules followed during the self-organization process. Since the inception of this research program in August 2003, we have sought to identify these rules and to apply them to synthetic problems. "Subcomponent self-assembly"¹ describes systems in which simple amines and aldehydes may be induced to come together around metal-ion templates *via* the formation of imine (C=N) bonds.^{2,3} We have thus been able to create complex structures such as a catenane (below, right)⁴ or a pair of helical molecules linked at a right angle through metal coordination (below, left), having a tertiary structure similar to that of a protein. These structures are capable of dynamic reassembly at both N→Metal and C=N linkages.⁵

1. J.R. Nitschke *Acc. Chem. Res.* **2007**, 40, 1032. J.R. Nitschke, M. Hutin, G. Bernardinelli *Angew. Chem. Int. Ed.* **2004**, 43, 67243. M. Hutin, G. Bernardinelli, J.R. Nitschke *Proc. Natl. Acad. Sci. USA* **2006**, 103, 176554. M. Hutin, C.A. Schalley, G. Bernardinelli, J.R. Nitschke *Chem. Eur. J.* **2006**, 12, 40695. D. Schultz, J.R. Nitschke *Proc. Natl. Acad. Sci. USA* **2005**, 102, 11191

Analytical Chemistry

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"Snapshots" of a heterogeneous catalyst at work: From integral towards spatially resolved spectroscopic monitoring of solid materials

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To gain information on the structure of solid materials such as heterogeneous catalysts, *in situ* X-ray absorption spectroscopy (XAS) is a well-suited technique. In certain processes the structure of a catalyst may vary along a catalytic reactor particularly if prominent gradients in temperature or concentration occur. The investigation of these phenomena requires spatially resolved techniques on a microscale.

Here we present structural data of noble metal catalysts during partial oxidation of methane (CPO). To efficiently record XAS spectra in a locally resolved way, a new approach is suggested using a CCD-camera [1]. The results are compared to those from μ XAS-measurements. In addition, the temperature gradient was determined using an infrared camera [2]. A correlation between the structure of the catalyst, the catalytic data and the axial temperature profile in the catalytic reactor was established. Tremendous structural changes of the noble metal particles within a gradient of less than 100 mm thickness and a strong dependence of the gradient on the reaction conditions (temperature, space velocity) were observed. Very recently, "snapshots" were not only taken of the variation in catalyst structure and temperature, but we even succeeded in following these changes in a time-resolved manner using a CCD- and an IR-camera.

[1] J.-D. Grunwaldt, S. Hannemann, C.G. Schroer, A. Baiker, *J. Phys. Chem. Lett.* **2006**, 17, 8674; J.-D. Grunwaldt, S. Hannemann, A. Baiker, P. Boye, C.G. Schroer, *Highlights of Analytical Chemistry*, *Chimia* **2006**, 60, 709.

[2] S. Hannemann, B. Kimmerle, J.-D. Grunwaldt, A. Baiker, P. Boye, C.G. Schroer, in preparation.

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Photoinduced functions in multicomponent molecular systems

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Light-induced processes are at the basis of fundamental natural phenomena as well as of a variety of applications. Since the functions that can arise from the interaction between light and matter depend on the degree of complexity and organization of the receiving 'matter', the research on these processes has progressively moved from molecular to supramolecular (multicomponent) systems. Examples of multicomponent systems capable to perform specific functions under light stimulation (photochemical molecular devices, PMDs) have been developed [1], relying on processes such as photoisomerization and photoinduced electron or proton transfer.

Here we will describe a few recent examples of PMDs studied in our laboratories [2], designed to (i) process binary information (molecular logic gates and circuits) or (ii) undergo controllable motions of some molecular components with respect to the others (molecular machines).

Apart from futuristic applications, investigations on PMDs can increase the basic understanding of a variety of processes, as well as develop reliable theoretical models. This research has also the important merit of stimulating the ingenuity of chemists, thereby instilling new life into chemical sciences.

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Chemometric tools to simplify method development: screening of doping agents in urinary samples.

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Analyses in anti-doping control occur in two steps. First, a generic fast screening analysis is used to determine the presence of a large number of forbidden compounds in urine. Second, in the case of positive result, a specific procedure (confirmatory analysis) has to be performed to quantify the substance(s).

Screening method development is tedious and time consuming due to the necessity to optimize the sample preparation of a large quantity of compounds. The use of chemometric tools is therefore proposed to reduce the number of tested analytes in method development by choosing representative compounds of the whole set.

In this study, a group of thirty-six doping agents consisting of diuretics and beta-blockers was used. A standard solution containing all analytes was loaded, washed and eluted with four different SPE sorbents. All fractions were collected and each compound was retrieved by LC-ESI-MS. In order to bring out differences among compounds, a multivariate analysis approach was used. A small number of groups emerged and subsequent method development was performed by selecting representative compounds in the obtained clusterisation.