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Combinatorial approach to design highly active and selective supported gold catalysts for PROX application

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Introduction

The selective oxidation of CO in the presence of hydrogen (PROX reaction) is one of most intensively studied reactions used in hydrogen technology [1].

Recent years the high-throughput and combinatorial approaches has obtained both great scientific and practical interest in catalysis research. In the last ten years method of catalyst library optimization improved very much. Different approaches, such as DOE, Genetic algorithm, Taboo search, Artificial Neural Networks, Holographic Research Strategy, have been applied by various research groups.

In PROX applications the main task is to have highly active and selective catalysts working in the temperature range between 80-100 °C. In this study methods of "real" and "virtual" combinatorial testing has been applied. Our method is based on a very fast optimization method, the Holographic Research Strategy (HRS) [3] and on the use of Artificial Neural Networks (ANNs), as an information mining tool [4]. The combination of these two methods provided a very fast informatic platform for catalyst library design by the combination of "real" and "virtual" experiments [5].

Materials and Methods

In the process of design of catalyst libraries the following approaches have been applied: (i) in the given library only one support (MgO) is used; (ii) gold is considered as an active phase, (iii) Pb was chosen as potential alloying components for Au; elements Ce, La and Y were chosen as compounds increasing the oxygen storage capacity of the support, while elements Sm and V were used as structural modifiers of the support. The Au_{max} value was limited to 4 wt %. All elements in the library were used in four – six concentration levels resulting in an experimental space containing about 25.000 - 35.000 experiments in each library. First the support was modified with the precursor compounds of elements A-G using a liquid dispensing robot. After washing and drying $HAuCl_4$ was introduced using the urea deposition method.

After reduction at 350 °C the catalysts were tested and optimized at 80 °C in a high-throughput reactor. An objective function (OF) was been created to rank the catalyst based on their conversion and selectivity values measured after 6 hours of time-on-stream. Alternatively, all catalysts were also treated using a combinative pre-treatment procedure. Results obtained in both treatment processes were evaluated by Artificial Neural Networks. ANNs provided an exact relationship between the composition and performance of catalysts after both pretreatment processes (reductive and combinative treatment, respectively). Based on ANNs and Holographic Mapping benchmark visualization of the whole experimental space was also performed.

Results and Discussion

The best hits in each generation are given in Table 1. The results unambiguously show the strength of our optimization tools. Table 2 shows the compositional differences after five generation using reductive and combined catalyst pretreatments. Combined reductive pretreatment resulted in a cheaper catalyst composition without alteration of the overall performance.

Table 1. Summary of results obtained in the 1-4 catalyst generations and after information mining using ANNs (reductive pretreatment).

Gene-rations	Composition w/w % of MgO support							Conver-sion %	Selec-tivity %	OF2*
	Au	Pb	V	Y	La	Sm	Ce			
0	4.0	3.0	-	-	-	-	-	79.2	40.8	**48.0**
1	3.0	1.0	1.0	1.0	1.0	0.0	0.0	79.7	44,5	**50.7**
2	3.0	1.0	1.0	1.0	0.0	2.0	0.0	84.5	49.4	**59.3**
3	4.0	2.0	2.0	0.8	0.0	3.0	0.2	88.1	52.2	**64.4**
4	4.0	2.0	2.0	1.4	0.2	3.0	0.0	92.2	50.1	**64.8**
5	4.0	3.3	0.2	0.6	0.9	3.2	0.0	93.0	55.9	**70.4**

*OF1 =conversion + selectivity, OF2 = special disability function

Table 2. Comparision of the best hits after the fifth generation using two different pretreatment processes. Results of „virtual" catalytic experiments

Pretreat-ment	Au	Pb	V	Y	La	Sm	Ce	Conv. %	Sele. %	OF2
Reductive	3.9	3.5	0.0	0.7	0.8	3.5	0.3	88.5	59.8	**70,4**
Combined	2.6	3.5	2.4	0.0	0.0	2.7	0.0	86.1	62.4	**70.2**

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Significance

The results show that Holographic Research Strategy combined with Artificial Neural Networks is a powerful tool for catalyst library design. The library design can provide new catalyst compositions, what were known earlier. The results show also that the mode of pretreatment has a significant importance both on the activity and the composition.

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