Direct synthesis of hydrogen peroxide in a wall coated microreactor using Au-Pd catalyst

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Abstract title	DIRECT SYNTHESIS OF HYDROGEN PEROXIDE IN A WALL COATED MICROREACTOR USING AU-PD CATALYST
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The development of a direct synthesis method for hydrogen peroxide production on site in desirable concentrations is of significant interest since it is a key selective oxidant in fine chemical industry. An obvious drawback in the formation of hydrogen peroxide from H_2 and O_2 is the very wide explosive region, 5–96 vol%, in the H_2 /O_2 system. To avoid the potential hazards from direct contact between H_2 and O_2, microreactors have been proposed. Microreactors are therefore inherently safe. An Au-Pd catalyst is applied in the synthesis of hydrogen peroxide in a microreactor immobilized on a SiO_2 layer coated on walls of microchannel.

Silica capillaries 320 µm in diameter were coated using a gas displacement method. A dynamic coating procedure was performed by displacing plug of the coating slurry consisting of colloidal silica, fumed silica, silica gel and sodium silicate as a binder using N_2. The most critical aspect of coating is the maintenance of film regularity during and after displacement of coating fluid. The uniformity of the coating thickness is greatly affected by the ability to maintain coating parameters such as coating velocity, temperature and solvent evaporation.The silica layer coated on the reactor walls was functionalized with the active metal species by impregnation with aqueous solutions of $HAuCl_4$ and $PdCl_2$ or with stabilized Au-Pd nanoparticles synthesized using two-phase synthesis method in dichloromethane following the protocol described elsewhere [1]. Concentrations of metal precursors where chosen to give approximately 5 Wt% total loading of bimetallic catalyst varying Au/Pd molar ratio.

To elucidate the advantages of using microreactors for the H_2O_2 synthesis, catalytic testing was done in a conventional Titanium batch autoclave reactor and in the microreactor set-up using the same type of catalyst. Experiments performed using diluted H_2/O_2 mixtures in both reactor systems showed similar productivity and selectivity values in both reactors.

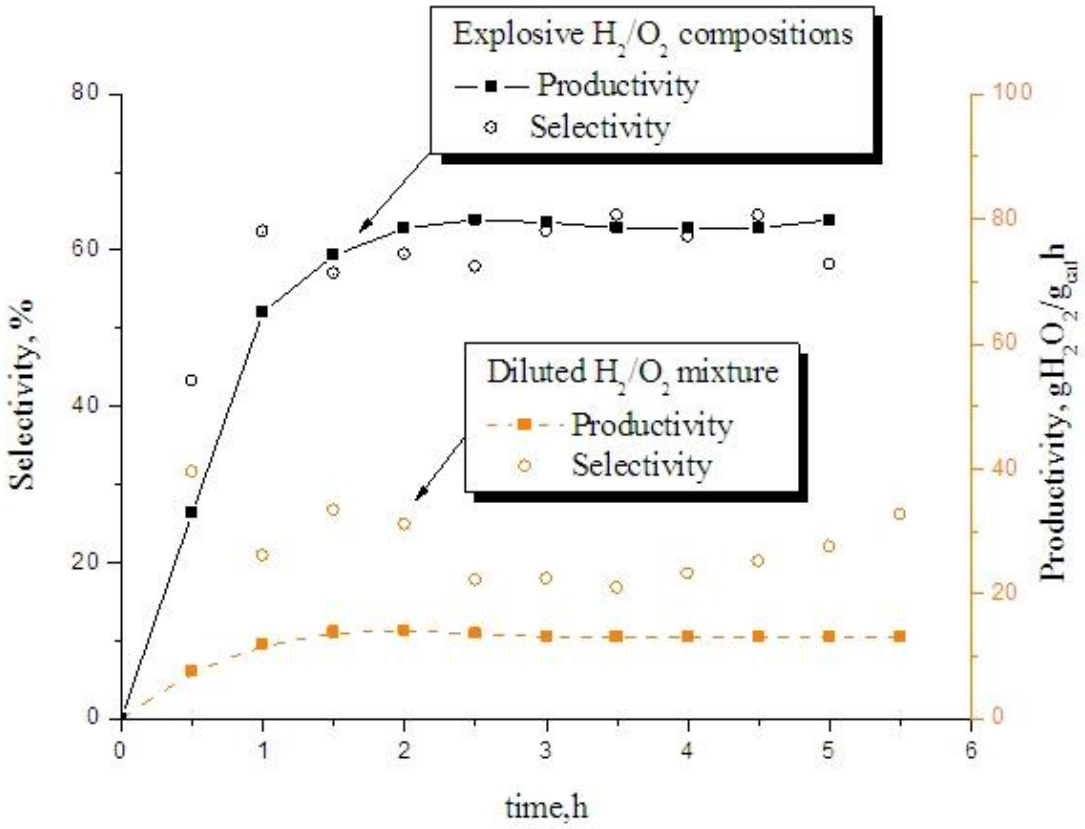
An important advantage of using microreactor technology is the possibility to use H_2/O_2 concentrations that would be considered explosive in traditional reactors. Figure 1 shows that switching the feed composition from non-explosive to explosive H_2/O_2 compositions results in a major increase in both the productivity as well as the selectivity while operating at 20 bars and 30 °C. The observed productivity of the Au-Pd alloy catalyst is almost an order of magnitude higher than both our observed productivity as well as reported values for similar Au-Pd catalysts under non-explosive conditions.

A comparison of the activities of the catalysts prepared using colloidal particles or directly impregnated in the capillary microreactore shows a superior activity for the nanoparticles. We will be presenting a comparison of the performance of these different types of catalysts as well present a kinetic study showing the benefits of operating with non-diluted H_2/O_2 mixtures.

Resulting process improvements regarding selectivity and productivities combined with enhanced process safety make microreactors the ideal reactor system for practical application in highly explosive reaction systems such as stoichiometric H_2/O_2 mixtures.

References

Serpell, C. J., Cookson, J., Ozkaya, D., Beer, P.D. Nat. Chem. 3, 478 (2011)



H2O2 synthesis using diluted vs. explosive H2/O2 mixtures in microreactor

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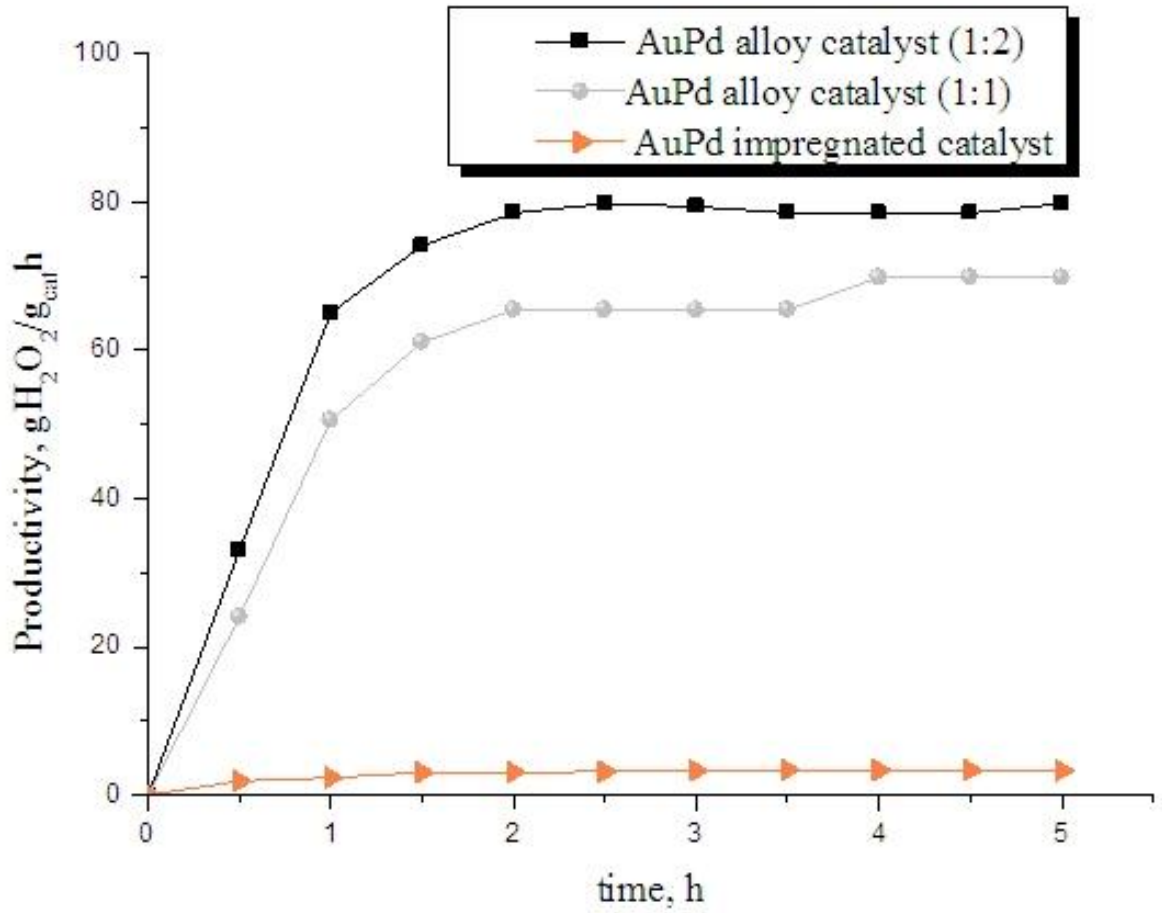

Productivities of a range of Au-Pd catalysts observed using explosive H2/O2 feed compositions