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Title:	Exploring Nanoplasmonics As a Tool for Surface Science
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Abstract:	<p>Bridging the structure and pressure gap in heterogeneous catalysis has proved to be the primary obstacle to the rational design of industrial catalysts. Nanoplasmonics-based sensing and spectroscopy provides a potential means to bridge the two gaps simultaneously. It allows the study of chemical interactions on nanoparticulate catalyst surfaces under in situ conditions of high temperature and pressure that mimic industrial conditions. The core of this experimental technique is the use of the plasmonic properties of sensor nanostructures (commonly gold) to probe changes on the surfaces of model catalyst-support films deposited on top of the sensing particles, using both refractive index sensitivity of peaks in the UV-visible spectra and surface enhancement of Raman signal for vibrational spectroscopy. There are three main aspects to the design of an experimental platform based on nanoplasmonics for the study of surface dynamics in heterogeneous catalysts; design and construction of suitable experimental apparatus such as a flow reactor, preparation of model catalyst-support architectures on the sensor surfaces, and the design of an optimized nanoplasmonic sensor. Here, all three aspects of this problem are addressed. Computational electrodynamics (FDTD method) is used to gain insight into the relationship of refractive index sensitivity, diffractive coupling, and near-field properties of arrays of gold nanocylinders on silica to particle morphology and array geometry. Two semi-analytical models, one based on an electrostatic eigenmode formalism and second based on a coupled dipole method, are proposed that can predict the variation of far-field absorbance properties of cylinders with morphology and variation in electric field enhancement with array geometry respectively. A novel thin film synthesis procedure to produce ultra-thin films of crystalline anatase on sensor surfaces at low temperatures is described which could, potentially, be generalized to a number of other oxide films. An ambient pressure flow reactor equipped with a UV-visible spectrometer and Raman spectrometer for detecting surface changes on modified nanosensors and a mass spectrometer for measuring composition of the gas flow has been constructed, and proof-of-concept experiments demonstrate its utility in monitoring gas-surface interactions on Au-anatase model catalyst on a lithographically fabricated nanoplasmonic sensor array.</p>
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