Title:	Surface Engineered Metal Oxide and Carbon Nitride Heterostructures for Photo-enhanced Electrochemical Ethanol Oxidation.
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Abstract:	The development of sustainable, clean and renewable sources of energy is required to mitigate the issue of rising demand of energy and depletion of fossil fuels. Recently, a rapid interest has been developed in the alcohol-based fuel cells as a promising alternative for clean power generation. Among the various kind of alcohols, ethanol is considered as the best alternative to be used in direct alcohol fuel cells due to its high energy density (~8 kWh kg -1), easy storage, safe transportation and environmental benign nature as it can be easily obtained from biomass. Along with its application in fuel cells, ethanol oxidation is widely used as a biomass oxidation process to produce commodity chemicals. It can also be coupled with hydrogen evolution reaction to replace sluggish oxygen evolution reaction in hybrid water electrolyzers to produce hydrogen which is a clean source of energy. Noble metal-based catalysts such as Pt and Pd are most frequently used in direct ethanol fuel cells but their high cost, scarcity and poisoning of the catalytic surface due to the adsorption of CO limit their applications. Therefore, non- precious metal based cost-effective, durable and highly efficient electrocatalysts need to be developed. The photo-assisted electrocatalysis can further accelerate the kinetics of the reaction and provide a more effective and renewable approach to convert solar and chemical energy to electrical energy. The work described in this thesis, utilizes heterogenous electrocatalysis process for the oxidation of ethanol molecules using metal oxide and carbon nitride based heterostructures. The developed heterostructures have extended light absorption in the visible region and thus can utilize solar energy for the photoelectrochemical process to achieve better efficiency. In the first work, a type II heterostructure of NiTiO 3 -TiO 2 has been synthesized via hydrothermal approach followed by calcination. The prepared heterostructures have been used for the electrooxidation of ethanol and show an enhancement of 5.5. % in the current density in presence of light. The better separation of charge carriers along with the formation of OH • assists in the oxidation of ethanol molecules at the catalyst surface. In the second work, a p-n junction based NiTiO 3 -NiO catalyst has been synthesized and the prepared heterostructure show better performance as compared to the pure NiO. An enhancement of 15% in the current density was observed on its use as a photoanode. In the third work, graphitic carbon nitride (gCN) based heterostructure has been synthesized using co-precipitation method. The effect of Co doping on the catalytic activity of gCN has been studied. The catalyst viiishows an enhancement of 85 % in the current density on photo illumination in the case of catalyst containing 3 % Co. The high electronegativity of N and oxophillic nature of Co results in overall better catalytic activity for ethanol oxidation. In the fourth work, the effect of metal chalcogenide incorporation in the gCN matrix has been tested. The catalyst was prepared by one step pyrolysis method and show good activity for ethanol oxidation resulting from the oxidation of the chalcogen atom leading to the formation of sulfates which changes the local coordination environment of the catalyst. In the last work, new strategy for the utilization of the plasmon induced charge separation in black gold (BG) decorated zirconia for efficient ethanol oxidation has been employed. The catalyst containing 6 mol % of black gold shows the best activity and enhanced photoelectrochemical activity due to the efficient charge separation at the BG-ZrO 2 heterointerface studied using ultrafast transient absorption spectroscopy. In summary, the present thesis focuses on developing cost effective and sustainable metal oxide and carbon nitride based heterostructures for their use as efficient catalyst for the photo/-electrochemical ethanol oxidation. The synthesized catalyst shows promising catalytic activity for the photo/-electrochemical ethanol oxidation.
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