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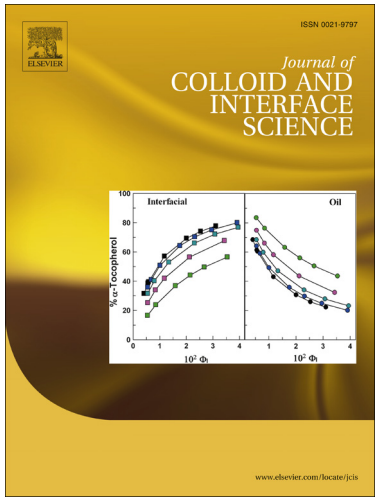

Pertinent parameters in photo-generation of electrons: comparative study of anatase-based nano-TiO_2 suspensions

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Pertinent parameters in photo-generation of electrons: comparative study of anatase-based nano-TiO_2 suspensions.

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Abstract

In the field of solar fuel cells, the development of efficient photo-converting semiconductors remains a major challenge. A rationale analysis of experimental photocatalytic results obtained with material in colloïdal suspensions is needed to access fundamental knowledge required to improve the design and properties of new materials. In this study, a simple system electron donor/nano-TiO_2 is considered and examined via spin scavenging electron paramagnetic resonance as well as a panel of analytical techniques (composition, optical spectroscopy and dynamic light scattering) for selected type of nano-TiO_2. Independent variables (pH, electron donor concentration and TiO_2 amount) have been varied and interdependent variables (aggregate size, aggregate surface vs. volume and acid/base groups distribution) are discussed. This work shows that reliable understanding involves thoughtful combination of interdependent parameters, whereas the specific surface area seems not a pertinent parameter. The conclusion emphasizes the difficulty to identify the key features of the mechanisms governing photocatalytic properties in nano-TiO_2.

Introduction

Since the beginning of mankind, the sun is the most abundant source of energy. Throughout centuries, attempts have been made to benefit from this free, and sustainable and renewable energy. In 1839, Becquerel discovered the photovoltaic effect allowing new route for solar energy conversion. However, both the high cost of solar energy conversion devices and the parallel massive exploitation of fossil fuel in the middle of the 20th century hampered the development of this field. As a consequence of successive breakdown events such as the oil crisis in the 70's, the public belief has accepted the concept of limited fossil resources. Since then, the continuous increase of energy needs and the more recent problem of global warming have brought solar energy to the scientific societal and economical forefront. In this context, numerous studies about the photoactivity of different oxide semiconductors have been performed since the second half of last century.[1] In 1972, Fujishima and Honda[2] reported the photoactivity of the titanium oxide (TiO_2) for the water photolysis process: an important pathway in the research field of clean, sustainable and renewable energy production. Today silicon technology is the main actor in the photovoltaic devices. In comparison, the TiO_2 semiconductor has the advantage of being cheaper, while displaying good stability in solution towards photocorrosion.[3] In addition to its great intrinsic properties TiO_2 is yet one of the most studied semiconductor in the field of solar energy and for many others applications ranging from water and air purification to self-cleaning surfaces.1 Abundant scientific reports have been focused on the photo-physicochemical properties of TiO_2, aiming at the rationalization of the semiconductor properties to optimize the performances of the material itself. Properties such as charge transfer, charge trapping, doping,[4] loading[5] or surface states have been investigated by varying parameters such as the scale of material (bulk or nanosized) and the crystallographic structure (anatase, rutile and brookite). In the case of nanostructured TiO_2, the size, shape (nanoparticle, nanorods, nanofibers[6]) and their specific surface area are currently under scrutiny. Despite the profusion of studies, a clear understanding of key parameters is not yet available. For instance, the photo-catalytic activity of the anatase phase compared to the rutile one is still debated.[7] In parallel, the photo catalytic efficiency of mixed phase rutile-anatase (e.g. commercial nano-TiO2 P25 from Degussa^{TM}) may be assigned to possible synergistic effects whereas other investigations conclude to the lack of particular features.[8] The complexity of theoretical modelling is further complicated when the chemical environment of the photocatalyst is taken into account. For instance, it has been shown that the photocatalytic reactivity of TiO_2 strongly depends on the target substrate to probe.[9] Finally, although theoretical approaches can yield conceptual approaches to the design of specific properties,[10,11,12] the design of operative complex configurations remains highly speculative. Hence, in the absence of a generally acknowledged well-defined physical statement, systematic studies are required for in depth studies of the photo-catalytic phenomena. In this context, our recently

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