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Abstract

Titanium dioxide (TiO_2) is a widespread metal oxide used in catalysis and photocatalysis, or coupled to non-thermal plasma for volatile organic compound (VOC) oxidation. Adsorption is a key step in the advanced heterogeneous oxidation processes; adsorption is partially addressed in that it is mainly investigated on fresh TiO_2 surfaces. However, the treatment of real effluents combines various pollutants with VOCs; among them, NOx are characterized by reactive adsorption properties on TiO_2 leading to irreversibly adsorbed species on the surface. The aim of this work is to determine the impact of NO_2 preliminary adsorption on the subsequent adsorption of a model VOC: acetaldehyde. Breakthrough methods are used to (i) characterize acetaldehyde adsorption on fresh and NO_2 covered TiO_2 surface, (ii) control the coverage of TiO_2 surface by irreversibly adsorbed $NO_x^-{}_{(ads)}$ species. In a first step, acetaldehyde adsorption on TiO_2 has been characterized. In a second step, acetaldehyde adsorption has been achieved on TiO_2 surface after NO_2 exposure corresponding to different surface coverages. Then, acetaldehyde adsorption parameters have been determined. Subsequently, it has been possible to plot the evolution of acetaldehyde reversibly and irreversibly adsorbed quantities, and reversible fraction adsorption constant, as a function of TiO_2 surface coverage by $NO_x^-{}_{(ads)}$ species, so called TiO_2 surface NO_2 ageing. Interestingly, both adsorbed acetaldehyde fractions are highly impacted by the presence of $NO_x^-{}_{(ads)}$ species on TiO_2 surface. The irreversibly adsorbed fraction is considerably decreased since the lowest values of TiO_2 surface coverage by $NO_x^-{}_{(ads)}$ species. Hypotheses related to competitive adsorption are not sufficient to explain the observed impact, suggesting that $NO_x^-{}_{(ads)}$ species modify TiO_2 surface chemistry and acetaldehyde reactive adsorption. The reversibly adsorbed fraction of acetaldehyde is highly impacted as well; reversibly adsorbed amounts are decreased since the lowest surface coverage by $NO_x^-{}_{(ads)}$ species. The corresponding adsorption constants are abruptly increased as soon as TiO_2 surface has been exposed to NO_2, suggesting the formation of a new adsorption mode for acetaldehyde on NO_2 exposed TiO_2 surface. This paper evidences the considerable impact of NO_2 on TiO_2 adsorption properties regarding VOCs. Considering that NOx may accumulate on TiO_2 the long term behaviour of such processes should be investigated further taking into account the NOx vs. VOC interaction for adsorption.

Keywords: adsorption; TiO_2; NO_2; acetaldehyde; surface coverage