

Public Abstract

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There is significant interest in methods to aid in the removal of carbon dioxide from our environment. The methods range from sequestration to catalytic transformations. In sequestration, CO₂ is collected from emission sources, and may be stored in a variety of materials including geological formations. Catalytic research often involves the transformation of CO₂ to another compound such as methane. The majority of these latter studies focus on the use of photocatalysis for the conversion of CO₂. An alternative method, discussed in this paper, is to examine the surface chemistry for the adsorption and thermal reduction of carbon dioxide with magnesium oxide and titanium oxide surfaces. We are also interested in gaining a better understanding of the refractory nature of CO₂.

The overall goal of these studies is to elucidate the surface reaction mechanism for the reaction of carbon dioxide on metal oxide surfaces with and without UV light. We combine the use of X-ray photoelectron and Auger electron spectroscopies to examine the surface chemistry of CO₂. First, carbon dioxide is exposed to the heated metal oxide surface and the surface atomic concentrations are measured after reaction. The substrate temperature and CO₂ exposure are varied in the experiment. Similar studies are also performed in the presence of UV light to gain a better understanding of photocatalysis.

Our results showed that the clean MgO surface exposed to different CO₂ (500-5000 Langmuir) exposure with a surface temperature (550-650 °C) can adsorb carbon on the surface. However, the exact structure of carbon is still unknown now. And the atomic concentration of oxygen on the very near surface decreased after exposure. Therefore, we can assume that the CO₂ adsorbs on the surface and forms some carbonate which then decomposes to other products, such as C and O₂. For the future experiments, we can use mass spectrometry to examine the gaseous products. Some studies have also shown that the interaction between CO₂ and MgO surface has an exchange of oxygen between the CO₂ and surface oxygen. We can use C¹⁸O₂ instead of C¹⁶O₂ and use mass spectrometry to verify what kind of gaseous products are generated; O₂, ¹⁸O₂, or O¹⁸O.

In photocatalytic reactions of CO₂ with TiO₂, when the clean surface exposed to different (CO₂) exposures at room temperature, there is an increasing atomic concentration of carbon on the surface. However, once the surface was under UV irradiation, the atomic concentration of carbon is decreased. At the same time, the oxygen concentration at the outermost surface layers increased. All of results indicated that the CO₂-exposed surface after UV light irradiation, resulted in some carbon leaving the surface as gaseous product, such as CO. Mass spectrometry can be used to identify the gaseous products. In order to prove whether there is an exchange of oxygen between CO₂ and the surface, we can use C¹⁸O₂ to perform the experiments instead of C¹⁶O₂, and then follow the mass spectrometry to determine the gaseous products, such as CO, C¹⁸O or some other products.

Future studies of metal oxide assisted catalytic reactions should also be extended to high surface area powdered surfaces. These high surface area powders can supply more sites for CO₂ activation. Modification to powdered samples by cation/anion doping or metal impregnation will serve as sites for CO₂ activation, and could lead to changes in the band gap. Alteration the band gap and increasing the number of active sites with powdered samples should help the development of processes that produce fuels and valued chemicals from CO₂ in the future.