Photon Stimulated Desorption of D_2O on the Surface of Oxide

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Abstract

Behavior of photon stimulated D_2O desorption on Cr_2O_3 have been studied with a Fourier transform infrared absorption spectrometer (FT-IR) and a quadrupole mass spectrometer (QMS). The light irradiation was found to enhance D_2O desorption. The 250 nm light stimulated D_2O desorption more strongly than the one of 400 or 600 nm. It seems that D_2O with a higher O-D wave number is more effective for light stimulated desorption.

1 Experimental

In order to observe the effect of photon irradiation on adsorption and desorption behavior, the quantity of desorbed D_2O was measured using a quadrupole mass spectrometer (QMS). Each sample set in the sample holder was irradiated with a light through the quartz glass window of a small chamber under a flow of the sweep gas. The volume of this chamber was 200 ml and the inner surface was gold plated. Sweep gas was N_2 added with D_2O or H2O vapor if necessary. Temperature of the sample was controlled in a range of 303 to 573 K. We also observed IR absorption under photon irradiation. Fig.1 shows the experimental apparatus schematically. After passing through the monochromator, the light was transmitted to the sample set in the diffuse reflectance apparatus. In these light irradiation experiments, Hg-Xe lamp was used as a light source. The relative spectral intensities of the light after through the monochromator were 1 (250 nm) : 5 (400 nm) : 3.5 (600 nm). The transmission factor of KBr window (2 mm thick) for the diffuse reflectance method was about 72 (250 nm) : 89 (400 nm) : 92 (600 nm).

2 Results and Discussion

Cr_2O_3 powder was contacted with N_2+D_2O at 41.3 Pa for 15 hours and then swept with N_2. After drying for a certain time (1400-3000 minutes), the light irradiation was started on the sample. The sweep gas containing desorbed species was led into a QMS in order to analyze the species with m/e=19 (HDO) and 20 (D_2O). Temperature was controlled to 453 K at the surface of the sample in order to effectively study adsorption and desorption of D_2O with low adsorption coverage. A typical desorption curve is shown in Fig.2. The light was injected after about 1400 minutes when no distinctive desorbed species were observed only by sweeping with N_2. The ordinate indicates the relative ion current intensity which was normalized to that of m/e=28 (sweep gas : N_2). These figures show that the light of 250 nm stimulated desorption of D_2O from the sample surface to a larger degree than the light of either 400 or 600 nm.

Observation of infrared absorption under photon irradiation were conducted by using the experimental system shown in Fig.1. Cr_2O_3 was contacted with Ar+D_2O at 43.7 Pa for about 15 hours, then switched from Ar+D_2O to Ar for drying. After about two hours, the light was injected to the sample. Fig.3 shows that the 250 nm light irradiation enhanced desorption of D_2O with a high wave number (2650 cm^{-1}). In the case of the light irradiation of either 400 or 600 nm, no desorption enhancement was observed within

experimental error. For D_2O with 2430 and 2510 cm^{-1}, no stimulated desorption was observed by the light irradiation.

From these experiments, it is summarized that

(i) 250 nm UV light irradiation was effective for desorption, but neither 400 nor 600 nm light.

(ii) D_2O with a higher O-D vibration frequency was easily stimulated by the ultraviolet light irradiation.

This means that a photon with a higher frequency may be more effective for stimulating desorption. One possible explanation for the mechanism of the light stimulated desorption is light energy acceptance by the solid surface and following energy transfer from the surface to the adsorbed species or adsorbing bond. These processes are considered to be effective for the adsorption system with low coverage which was the experimental condition in the present study. The experimental results seem to show that energy transfer for desorption is more effective for weakly bonded D_2O.

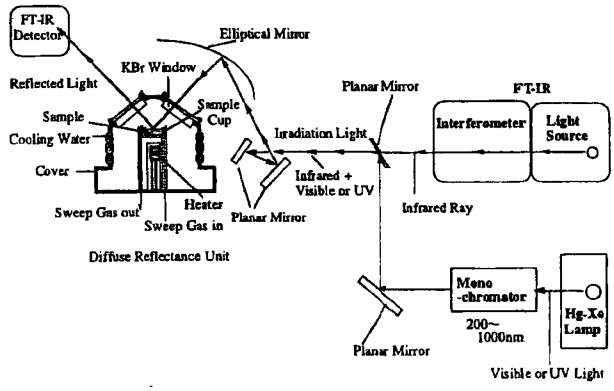

Fig.1: A schematic diagram of FT-IR diffuse reflectance method with simultaneous photon injection.

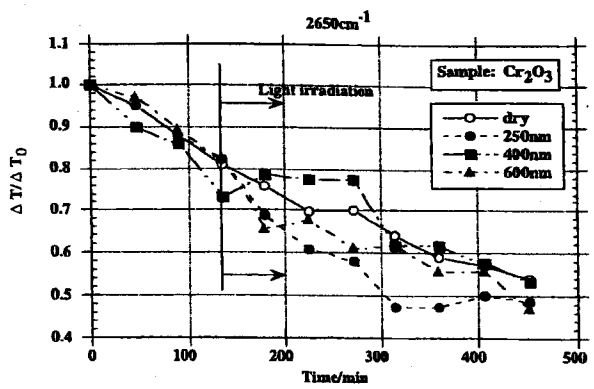

Fig.3: Change of absorption spectra for Cr_2O_3 during D_2O desorption under photon irradiation at 453 K. ΔT is normalized by that $\Delta Tt = 0$.

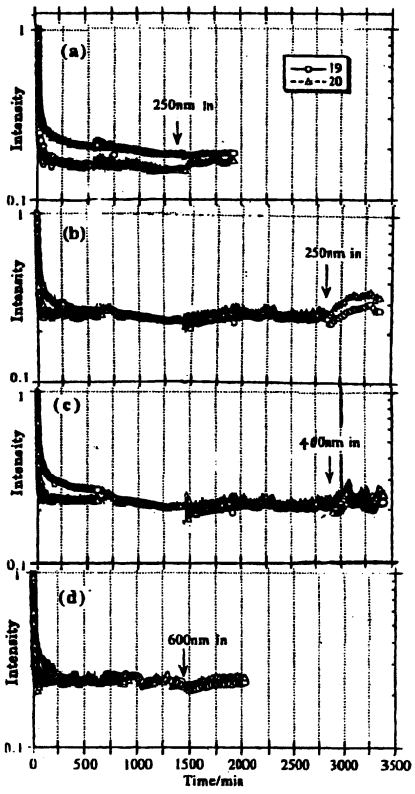

Fig.2: Change of desorption intensities for the mass numbers 19 and 20 from Cr_2O_3 at 453K. (a) Blank test without specimen, (b) 250nm, (c) 400nm, (d) 600nm.