Changes in Catalytic Activities of Cu Thin Films by Surface Acoustic Waves propagated on Ferroelectric $LiNbO_3$

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A surface acoustic wave was effective for the activation of a Cu thin film catalyst.

The propagation of a surface acoustic wave (SAW) on ferroelectrics is one of the most interesting surface phenomena, since the wave leads not only to the displacement of atom particles near the surface but also to the generation of an associated electric field.[1,2] It would be expected that SAW has a high potential to enhance the function of catalytic surfaces, but there have so far been few studies. In the present work, SAW has first been employed for a metal thin film as catalyst, and we have found that it has a dynamic effect on the catalysis.

A single crystal of ferroelectric $LiNbO_3$ with x-axis propagation (128° rotated Y-cut) was used as a SAW substrate. The sets of interdigital transducer (IDT) electrodes were microlithographically fabricated on both the ends of the substrate. The rf-generating IDT electrode had 20 pairs of double fingers with a distance of 100 μm at each pair. The receiving electrode was the same except for 14 double fingers. The IDT electrodes were composed of a 10 nm Cr layer covered with a catalytically inactive Au layer of 100 nm. The two sets of the IDT electrodes were 16 mm apart, in the middle of which catalytically active Cu films as thick as 10 nm were deposited over a geometric area of 1 cm^2. The rf-signals were generated by a network analyser, amplified and then applied to the thin Cu catalyst. The output signals through the catalyst phase were returned to the network analyser. The band path frequency of SAW observed was 19.5 MHz and had a width of 1.5 MHz at 20 dB loss. This centre frequency was consistent with a calculated value, 19.9 MHz. The gaseous catalytic reactions

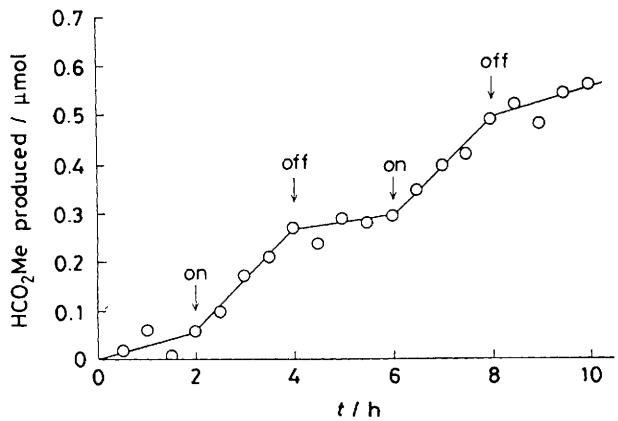

Figure 1. Changes in catalytic activities with SAW-on and -off for the dimerization of formaldehyde. Reaction temperature; 333 K, pressure of formaldehyde; 0.67 kPa.

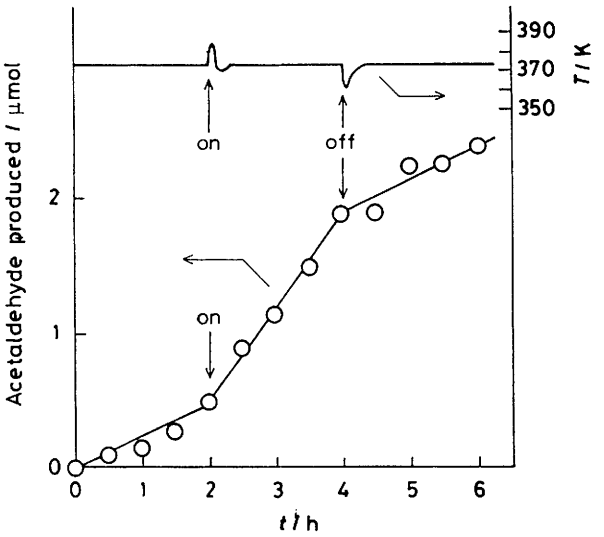

Figure 2. SAW effects upon the catalytic activities and surface temperature. Reaction; acetaldehyde formation from ethyl alcohol (4 kPa) and oxygen (4 kPa).

such as the dimerization of formaldehyde and the oxidation of ethyl alcohol were carried out in a conventional high vacuum apparatus with a circulation system.

Figure 1 shows the results on the dimerization of formaldehyde to methyl formate. When SAW was applied, the rate of methyl formate production immediately increased and remained at a high constant level. With SAW-off, the rate decreased to almost the same level before SAW-on. It is interesting to note that the SAW effect was reproducible for the repetition of on and off cycles and enhanced the activity by a factor of 3.5. Figure 2 shows the SAW effect upon the oxidation of ethyl alcohol to produce acetaldehyde. The generation of SAW leads to an increase in the activity by a factor of 2.7 and the high activity continued while SAW was being operated. The original low activity was obtained by turning SAW off.

With respect to the increases in the catalytic activities, one might consider the influences of changes in the temperature which are caused by SAW power. The temperature of the catalyst surface was carefully monitored by a thin CA thermocouple which was brought into direct contact with the surface. Upon SAW-on, an initial increase in temperatures from 373 to 385 K occurred (Figure 2); the surface temperatures remained higher than 373 K for 8 min, then decreased and recovered to the original level within 12 min under the control of an external heating system. On the other hand, the higher activities continued over 2 h as long as SAW power was applied. Therefore, these findings indicate that the effect of SAW is not based on thermal contribution but it has a role to activate the deposited catalytic phase.

SAW is able to cause the displacement of lattice atoms and a high electric field. It is likely that SAW is effective for the catalytic reactions involving polar molecules, because it has no effect in the ethylene hydrogenation.[3] A previous study on a thin Cu film deposited on the (+) and (−) polar z-$LiNbO_3$ showed that the oxidation of methyl alcohol was promoted by the (−) polar than by the (+) polar substrate.[4] This indicates the important role of an electric field. Thus, the generation of a high electric field by SAW seems to be responsible for the activation.

The interesting role of SAW was evidenced by the present work, and it is expected that SAW becomes an external electric signal to develop the function of a catalytic phase.

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References

1 R. M. White, *Proc. IEEE*, 1970, **58**, 1238.
2 H. Wohltjen, *Sensors Actuators*, 1984, **5**, 307.
3 Y. Inoue, M. Matsukawa, and K. Sato, *J. Am. Chem. Soc.*, in the press.
4 Y. Inoue, T. Imai, and K. Sato, *Chem. Phys. Lett.*, 1989, **160**, 118.