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Rapid evaluation of oxidation catalysis by gas sensor system: total oxidation, oxidative dehydrogenation, and selective oxidation over metal oxide catalysts

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Abstract

The rapid evaluation of catalysis is an indispensable technology for the success of combinatorial chemistry. A small-sized, less expensive, easily operating screening is desirable for parallel settings which dramatically shortens the evaluation time. Recent advances in gas sensors have enabled us to use them for the rapid evaluation of oxidation catalysis. Three typical catalytic oxidations over metal oxide catalysts were evaluated by gas sensor systems optimized for each catalytic system. The first one is the total oxidation of carbon monoxide in air. Five catalytic combustion-type gas sensors were used in a parallel reactor system to shorten the evaluation time. The second one is the oxidative dehydrogenation (ODH) of ethane over the mixed oxide of nickel and iron. The evaluation of the ODH catalysis was performed by a selective olefin sensor which determines the concentration of C_2H_4 in C_2H_6. The third one is the selective oxidation catalysis of propane over alkali modified Fe/SiO_2. The effluents including CO, CO_2, aldehydes and ketones in propane were analyzed by the CO, CO_2 and semiconductor-type gas sensors selective toward aldehydes and ketones. These evaluation results indicated that gas sensors have a good potential for the rapid evaluation of oxidation catalysts.

Keywords: Combinatorial chemistry; Rapid evaluation; Gas sensor; Oxidation catalysis

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1. Introduction

The success of the combinatorial approach in pharmaceutical chemistry encourages chemists who study other functional materials including catalysts to apply the combinatorial approach to their areas. The combinatorial approach requires at least three basic technologies. The first one is the parallel synthesis of many hopeful candidates. The second one is their evaluation in a short time. The third one is the optimization of a hopeful candidate and planning an improved library. Although the problems depend on each catalytic system for the application of the combinatorial method, the development of a rapid screening would be the key technology in many catalytic systems.

Several publications have dealt with the evaluation technology in heterogeneous catalysis [1–6]. IR thermography was used for the evaluation of hydrogen oxidation catalysts placed two-dimensionally in order [7]. This method is suitable for the investigation of catalysts for highly exothermic or endothermic reactions. The laser-induced resonance enhanced multiphoton ionization (REMPI) is a powerful tool for the very high throughput screening of the dehydrogenation of cyclohexane [8,9]. Cong et al. [10,11] utilized

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mass spectroscopy for the evaluation of CO oxidation, NO reduction or ethane oxidative dehydrogenation (ODH) catalysts. Hoffmann et al. [12] used a non-dispersive infrared (NDIR) sensor to evaluate the CO oxidation catalysis. It is necessary to apply the combinatorial chemistry to develop a new rapid evaluation method that is informative about the reaction products. Especially, an evaluation module of small size is suitable for parallel work order to dramatically shorten the evaluation time.

The identification of gaseous compounds has recently made remarkable progress using gas sensors. There are several types of gas sensors: the catalytic combustion type, semiconductor type, potentiometric type, NDIR type, and so on [13]. Gas sensors of the first two types are remarkable in their conciseness. Sintered ceramic beads of about 0.5 mm diameter are usually used as the sensing elements of these sensors. Such a small size will enable us to use the sensors inside miniature reactors specially designed for the high throughput screening. Although the gas selectivity of the sensors is not better than that of gas chromatography or other conventional analyses, these small sensors respond quickly and also continuous observation of an effluent is available.

In this study, we have demonstrated the potentiality of gas sensors for the rapid evaluation of heterogeneous catalysts for three kinds of oxidations, the simple combustion of CO, the ODH of ethane, and the selective oxidation of propane to oxygenates. The catalysts for the CO oxidation are required for many aspects. CO is the origin of air pollution and is highly toxic so that its removal is important for our respiration. The CO oxidation catalysts are also required for the CO_2 laser application [14] and in fuel cell technology [15]. The ODH at lower temperature is of considerable interest for practical reasons, especially, ethane ODH is important for ethylene production. The selective oxidation of the lower alkanes into oxygenates is also important for the utilization of natural gas.

The examined sensor system and catalysts are as follows. The catalytic combustion type sensor for flammable gases was examined for the evaluation of supported Rh catalysts in the CO oxidation. An array of five sensors was also attempted for use in a parallel reactor system. A semiconductor SnO_2 sensor selective to olefins was employed for the evaluation of the ethane ODH on ferrite catalysts, which have been studied for the ODH of butene [16] and butane [17]. A combination of a semiconductor oxygenate selective sensor, a potentiometric CO sensor and an NDIR CO_2 sensor was used to determine the selectivity of oxygenates as well as their total yields during the oxygenate formation from propane over aklali-iron-silica catalysts [18–22]. The obtained results with gas sensors were compared with those from gas chromatography. Here we show the potential of the gas sensors for the rapid evaluation of oxidation catalysts.

2. Experimental

2.1. Gas sensors

A catalytic combustion-type sensor for the detection of flammable gases was used to determine the CO concentration. An alumina bead of about 0.5 mm diameter, on which platinum is loaded as a catalyst, is the sensing element of this sensor. The coiled wire of a Pt–Rh alloy is placed inside the alumina bead to maintain the element temperature at about 500°C using an electric current. The catalytic combustion of flammable gases, such as CO increases the temperature of the sensing element in proportion to the concentration of the flammable gases when sufficient oxygen is contained in the atmosphere. The change in the bead temperature is measured as a change in the resistance of the Pt–Rh wire. The combustion heat of the flammable gases determines the output signal from the sensor without gas selectivity. The range of the detectable gas concentration is from 0.01 to 10% in air.

The semiconductor-type gas sensors were employed for the analysis of the effluents including olefins or oxygenates. An 8-channel SnO_2 sensor array deposited on an 1.0 cm × 1.0 cm alumina plate was fabricated [23]. Flammable gases bring about changes in the resistance of SnO_2 when the sensor temperature is kept at 600°C. In order to improve the gas selectivity, seven kinds of catalyst layers, 13 wt.% SiO_2/Al_2O_3, 28 wt.% SiO_2/Al_2O_3, ZnO, TiO_2, ZrO_2, 3 wt.% Mo/SiO_2, 3 wt.% V/SiO_2, were deposited on seven SnO_2 sensors of the array [24]. Through a preliminary survey, the SnO_2 element covered with 28 wt.% SiO_2/Al_2O_3 was selected as selective olefin sensor and SnO_2 element covered with TiO_2

or 13 wt.% SiO_2/Al_2O_3 was used as the selective oxygenate sensors.

A combination of three sensors (two semiconductor-type sensors selective to oxygenates and a potentiometric CO sensor) and NDIR sensor (LX-720 CO_2 analyzer, Iijima electronics corporation) was used for the evaluation of the catalysis for the selective propane oxidation. The semiconductor-type sensors (oxygeante sensors-1 and -2) consist of SnO_2 sensitized by TiO_2 or by 13 wt.% SiO_2/Al_2O_3, respectively. Each sensor is placed in a microchamber and connected in series to each other by tubes. A micro-pump is placed at the end of the tube. The output signal of each gas sensor was checked with air including an ambient amount of CO, CO_2, C_2H_4 or formaldehyde once a week. No significant change was observed in the output signals of these sensors for 1 month.

2.2. Catalyst preparation

Silica was purchased from the Merck KGaA (BET surface area = 400 $m^2\,g^{-1}$). Fumed titania (P-25) was obtained from Nippon aerosil. Tungsten(VI) oxide, tin(IV) oxide, ceria, rhodium(III) nitrate and tris(acetylacetonato)iron(III) were purchased from Kishida Reagents Chemicals. Rhodium nitrate was purchased from Mitsuwa Chemicals. Acetonitrile was obtained from Kanto Chemicals. These reagents were used without any other purification. Water was deionized by an ion exchange resin and distilled before use.

The catalyst used for the CO oxidation was prepared by the impregnation method. An aqueous solution (50 cm^3) of rhodium nitrate was slowly added to MO_x (1 g, Rh/M = 1/100, M = Sn, W, Si, Ti, Ce). The excess water was removed under reduced pressure at 50°C and dried at 70°C under atmospheric pressure overnight. The catalysts were calcined at 400°C in air for 3 h. Each catalyst was reduced by 5 vol.% H_2 in He at 350°C for 3 h and cooled to 30°C under nitrogen before the catalysis measurement.

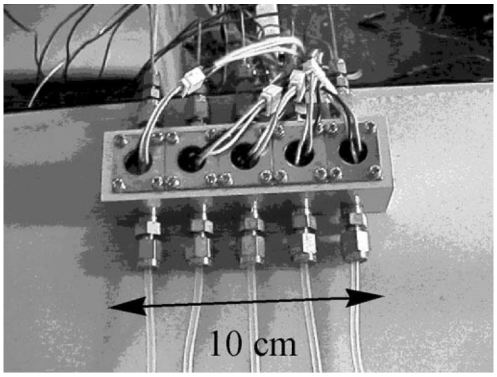
The mixed oxides of nickel and iron used for the ODH were prepared by co-precipitation from metal nitrates and potassium carbonate solutions. An aqueous solution (300 ml) of nickel nitrate hexahydrate (2.91 g, 0.010 mol) and iron nitrate nonahydrate (8.08 g, 0.020 mol) was slowly added to an aqueous solution (200 ml) of potassium carbonate (7.00 g, 0.051 mol) with vigorous stirring. A large quantity of dark brown precipitates immediately appeared. After stirring several hours, the precipitates were filtered and washed with 3 l of distilled water, dried under vacuum at room temperature, and finally calcined at 400°C in air for 3 h.

A series of silica samples with isolated iron was used as the selective oxidation catalysts for propane. Iron-loaded SiO_2 catalysts were prepared by the incipient wetness method. Iron(III) acetylacetonate (8.80 mg, 2.50×10^{-2} mmol) was dissolved in 3 cm^3 of acetonitrile. The solution was slowly added to silica (3.00 g, 5.00×10^{-2} mol) and placed on a microwave soniator for 15 min. After drying at 60–80°C overnight in air, the silica was calcined at 600°C for 5 h at a heating rate of 10°C min^{-1}. Alkali modified Fe/SiO_2 catalysts were prepared with the nitrate salts of each alkali metal and Fe/SiO_2. An aqueous solution (1.00 ml) of alkali nitrate (0.15 M) was added to 1.00 g of Fe/SiO_2. The conditions for drying and calcination were the same as those of Fe/SiO_2. The BET surface areas of the catalysts were between 330 and 360 $m^2\,g^{-1}$ after calcination.

2.3. Reaction conditions

Each Rh/MO_x (150 mg) catalyst was loaded into a quartz tube reactor (10 mm i.d.). Air including 1 vol.% CO at a pressure of 101 kPa was passed through the reactor at a flow rate of 50 $cm^3\,min^{-1}$ (SV = 20,000 $h^{-1}\,ml\,g^{-1}$). Each catalysis of five samples was measured at the same time. The reaction temperature was increased from 30 to 150°C for 2 h (ramp rate = 2°C min^{-1}) in an oven. The effluents from the reactors were analyzed by the 5-channel gas sensor system shown in Scheme 1.

The oxidative dehydrogenation reaction (ODH) of ethane was also carried out under atmospheric pressure. The catalysts were pretreated with flowing air (50 ml min^{-1}) at 600°C for 30 min. The mixture of C_2H_6, N_2 and O_2 (42:60:18, 120 ml min^{-1}) was then passed through the 300 mg of catalysts diluted with 2.7 g of silica sand in the micro-fixed bed reactor (i.d. = 10 mm) at ambient temperature (SV = 20,000 $h^{-1}\,ml\,g^{-1}$). The effluent diluted with 880 $cm^3\,min^{-1}$ of dried air was analyzed by the 8-channel gas sensor system. The effluent was introduced into the sensor system for 2 min and the output signals were recorded.



Scheme 1. Five parallel catalytic combustion type sensors for the evaluation of CO oxidation catalysis.

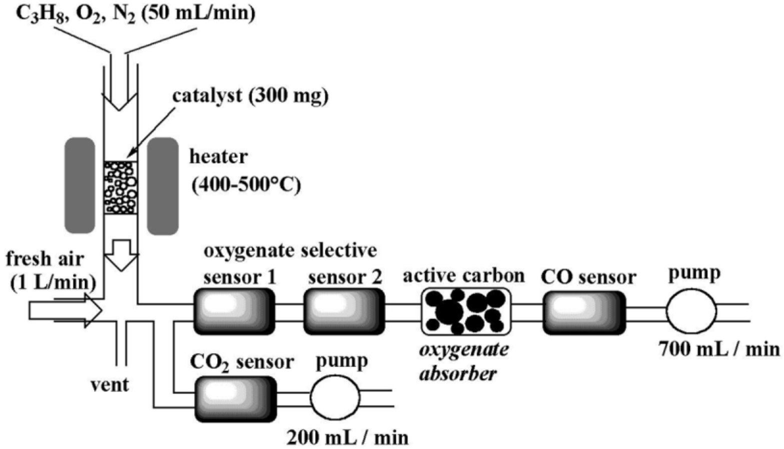
Each of the alkali/Fe/SiO_2 (300 mg) samples was loaded into a quartz tube reactor (10 mm i.d.). The catalysts were pretreated at 600°C for 30 min in flowing air (50 cm^3 min^{-1}) before the catalysis measurements. A mixture of propane, oxygen and nitrogen in the ratio of C_3H_8:O_2:N_2 = 20:16:64 at a pressure of 101 kPa was passed through the reactor at a flow rate of 50 cm^3 min^{-1} (SV = 10,000 h^{-1} ml g^{-1}). The effluent from the reactor was analyzed by the gas sensor system shown in Scheme 2. The gas from the catalytic reactor was diluted with dry air to obtain the optimal concentration for each sensor and then introduced into the inlet of the gas sensors.

3. Results and discussion

3.1. Evaluation of CO oxidation catalysis with five paralell sensors

The oxidation of 1 vol.% CO in air over Rh/TiO_2 was carried out under atmospheric pressure from 30 to 150°C. The heating rate of the temperature was 2°C min^{-1}. The CO concentration of an effluent from the reactor was continuously determined by the CO gas sensor and gas chromatograph every 10 min. Fig. 1 shows the output signal and CO conversion determined by the CO gas sensor and CO conversion by FID–GC on the CO oxidation over Rh/TiO_2. This indicates that the CO concentration decreased with increasing temperature, i.e. the CO conversion rate increased at higher temperature. The closed circles in Fig. 1 indicate the CO conversion determined by gas chromatography. The conversion curve calculated from the output signals of the CO gas sensor are denoted by the closed circles. This result clearly shows that the evaluation result with the CO sensor is as same as that with the gas chromatograph.

CO oxidation catalysis of a series of Rh/MO_x was evaluated by five CO gas sensors at the same time. The evaluation time for each sample became 1/5 compared with the time for one method. The reaction tubes were placed together in the oven to control the reaction temperature. Fig. 2 shows the output signals from the five CO sensors in the temperature range



Scheme 2. Schematic drawing of gas sensor system for the evaluation of selective propane oxidation.

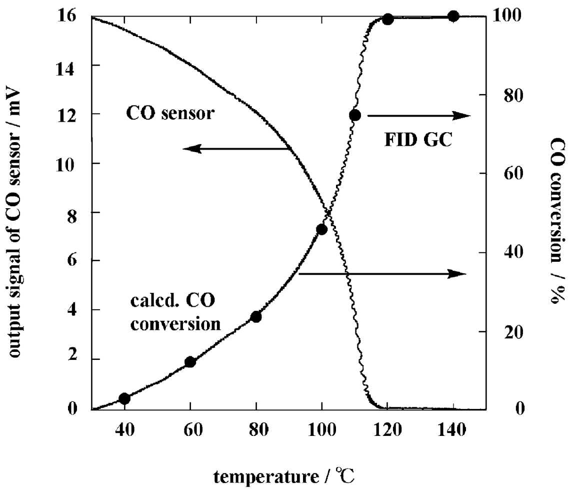


Fig. 1. Reaction temperature dependence of output signal from CO sensor on CO oxidation over 1 atm% Rh/TiO_2 and CO conversion determined by CO gas sensor and FID–GC.

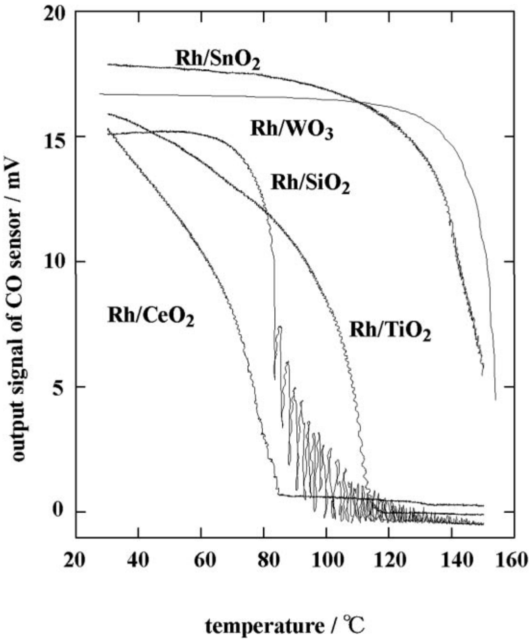


Fig. 2. Reaction temperature dependences of output signals from CO sensors on CO oxidation over 1 atm% Rh/TiO_2, SnO_2, SiO_2, WO_3, CeO_2.

from 30 to 150°C. The Rh/CeO_2 catalyst brought about a decrease in the CO concentration at the lowest temperature from among the supported Rh catalysts. The order of the reactivity, Rh/CeO_2 > Rh/SiO_2 > Rh/TiO_2 > Rh/SnO_2 > Rh/WO_3, is clearly demonstrated in Fig. 2. It is also worth noting that the gas sensor precisely monitors the oscillation of the CO oxidation on Rh/SiO_2. This continuous gas analysis is useful for the study of dynamic phenomena. Although the lack of gas selectivity is the drawback of a catalytic combustion-type sensor, gas selectivity is not required for the evaluation during simple combustion. When compared with a conventional single line flow reactor equipped with an automatic gas chromatograph, a higher throughput of more than 20 times was obtained for the five parallel reactors with five sensors.

3.2. Evaluation of ODH catalysis with olefin sensitive sensor

Fig. 3a shows the correlation between the output signals from the olefin selective sensor and the concentration of C_2H_6, C_2H_4 and CO in N_2. The output signal of 1–2 V was obtained in the presence of 1000–5000 ppm C_2H_4. The output signal of 0.5–1.2 V was obtained in the presence of 1–5 vol.% C_2H_6. The presence of CO did not affect the output signal. Although the sensor is more sensitive toward C_2H_4 than C_2H_6, the effect of C_2H_6 on the output signal is not negligible when the conversion of C_2H_6 is lower. Fig. 3b shows the relationship between the output signal of the sensor and the concentration of C_2H_4 in C_2H_6. The total flow rate of C_2H_4 and C_2H_6 was maintained at 42 ml min^{-1}. The gases were diluted with 958 ml min^{-1} of dry air before introducing the sensor system. The output signal increased in proportion to the concentration of C_2H_4 with a linear correlation. This result indicates that the gas sensor is useful for the determination of the C_2H_4 concentration and the evaluation of the ODH catalysis. The concentration of C_2H_4 in a reaction effluent was evaluated by the correlation curve.

The ethane ODH catalysis of the mixed oxides of nickel and iron were evaluated by both the gas sensor and an FID gas chromatograph. Fig. 4a shows the C_2H_4 yield determined by the gas sensor compared with that by FID–GC on ODH over $NiFeO_x$ in the

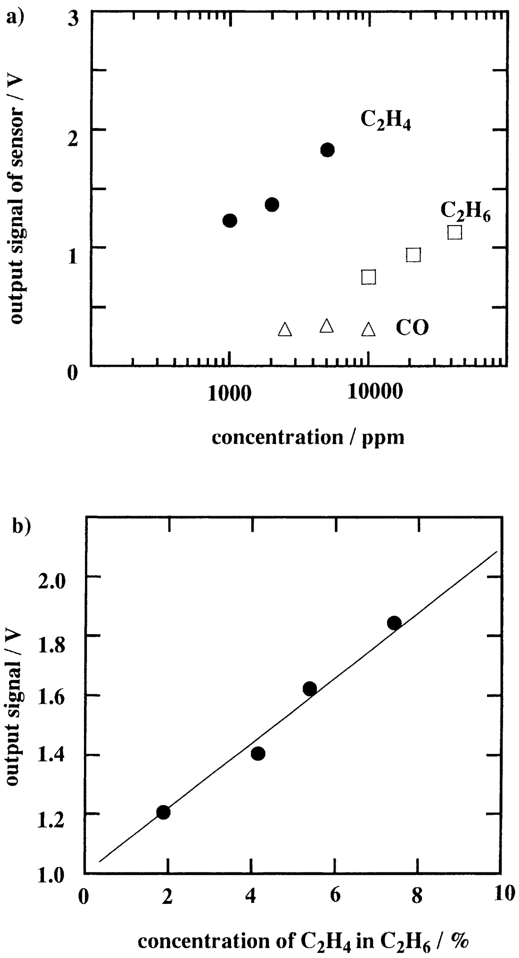


Fig. 3. (a) Output signals of selective olefin sensor toward C_2H_4, C_2H_6 and CO in air, and (b) output signals of selective olefin sensor toward C_2H_4 in C_2H_6.

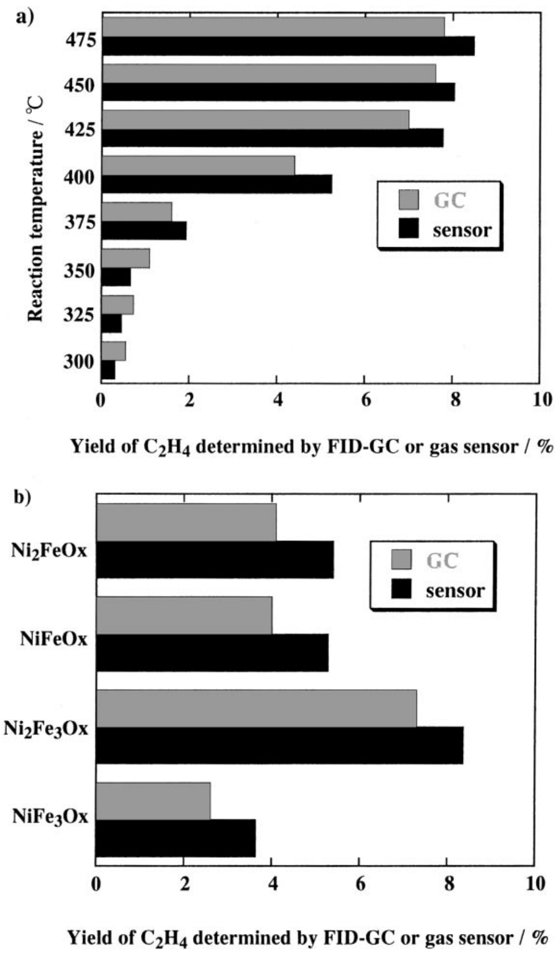


Fig. 4. (a) Evaluation of C_2H_4 yield by gas sensor and FID–GC on ethane ODH over $NiFeO_x$ from 300 to 500°C, and (b) evaluation of ethane ODH catalysis over Ni_2FeOx, $NiFeO_x$, $Ni_2Fe_3O_x$ and $NiFe_3O_x$ by selective olefin sensor and FID–GC.

temperature range from 300 to 500°C. The output signals of the gas sensor were reproducible in spite of the higher production of water vapor at high conversion. Although the gas sensor overestimated the C_2H_4 yields, the difference is less than 0.8 point and the tendency, i.e. high yield of C_2H_4 at high temperature, was correctly found. The gas sensor was useful to investigate the temperature-dependent catalysis of $NiFeO_x$. Fig. 4b shows the evaluation result of the mixed oxide of nickel and iron with different Ni/Fe ratios. The reaction was performed at 400°C. The ratio of Ni/Fe was changed ranging from 1/3 to 2/1:$NiFe_3O_x$, $NiFe_2Ox$, NiFeOx and Ni_2FeOx. The order of better catalysts estimated from the output signal of the gas sensor is the same as that estimated from the gas chromatograph. These results indicate that the gas sensor would be useful to optimize the ODH catalysis of the mixed oxides of nickel and iron in their composition and reaction temperature.

The sensor used here, SnO_2 covered with 28 wt.% SiO_2/Al_2O_3, is sensitive toward hydrogen and aldehydes as well as toward ethylene. Since neither hydrogen nor aldehyde is produced under the present reaction conditions, reliable yields of ethylene were obtained from the sensor output. When aldehydes are produced in the reaction, an adsorbent, such as active carbon filter is required in front of the sensor to remove aldehydes from the analyzed gas. A hydrogen sensor should also be necessary to compensate the output signal from the olefin selective sensor. In the next section, the combination of gas sensors is employed to evaluate the reaction selectivity during propane oxidation.

3.3. Evaluation of selective oxidation catalysis of alkali modified silica with iron ions

Usually, several or multiple kinds of compounds are formed as the products during the selective oxidation of propane. The formations of CO, CO_2, aldehydes, such as acrolein, and ketone were observed over iron-silica catalysts [19,20]. During the initial stage of the investigation of a new selective oxidation system, we are mainly interested in the conversion of propane and the selectivity toward useful oxygenate products as chemical resources. Here, the semiconductor-type gas sensors selective toward the oxygenate are employed in order to evaluate the propane oxidation products, with a combination of the potentiometric CO sensor and the NDIR CO_2 sensor [25].

Fig. 5a shows the response of the oxygenate sensor-1 (SnO_2 sensitized with TiO_2) toward an alcohol (1-propanol), aldehyde (propion aldehyde), ketone (acetone), carbon monooxide (CO), and propane. The sensor is sensitive to the alcohol, aldehyde, ketone but not sensitive to CO and propane. On the other hand, the oxygenate sensor-2 (SnO_2 sensitized with 13 wt.% SiO_2/Al_2O_3) is less sensitive to the alcohol than aldehydes as shown in Fig. 5b. Alcohol formation can be estimated from a comparison of the output signals of the oxygenate sensors-1 and -2.

Usually the conversion of propane is kept at less than 15% for the selective oxidation. The concentration of propane is nearly 100 times higher than those of CO and other oxygenates during the reaction gas at lower conversion. The sensors used here is selective enough to confirm the oxygenate formation in the reaction gas. The effects of produced water, hydrogen and olefin on the output signals of the oxygenate sensors-1 and -2 are negligible under the present reaction condition. As for the potentiometric CO sensor, it requires an active carbon filter as an absorber for propane and oxygenates which disturb the determination of the precise CO concentration. The selectivity of the NDIR CO_2 sensor is high enough for this purpose.

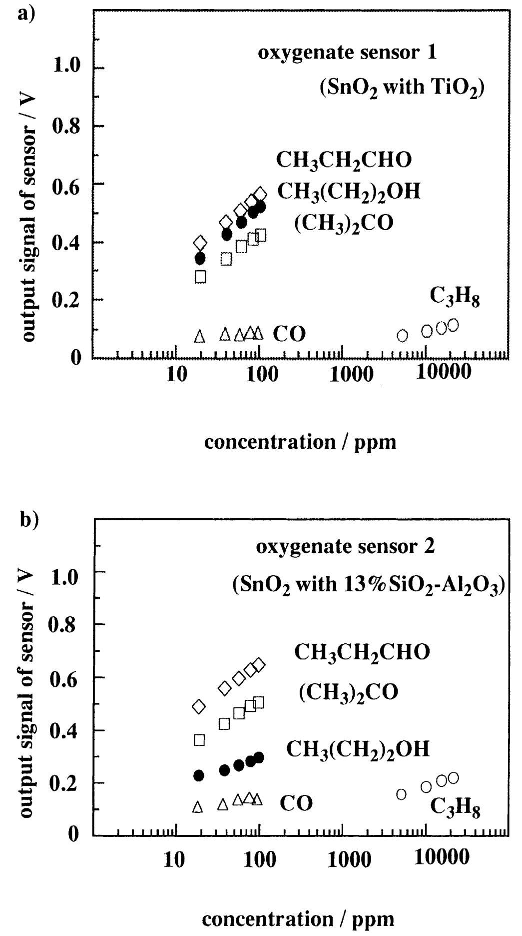


Fig. 5. Response of (a) oxygenate sensor-1 and (b) oxygenate sensor-2 toward aldehyde (CH_3CH_2CHO), alcohol ($CH_3(CH_2)_2OH$), acetone, CO and propane.

Fig. 6 shows the change in the output signals from the oxygenate sensors-1 and -2, the potentiometric CO sensor and the NDIR CO_2 sensor after the introduction of the reaction gas into the gas sensor system. The time required for the 90% response of each gas sensor is 20, 90 and 40 s for the oxygenate sensor, CO sensor and CO_2 sensor, respectively. The slower response of the CO sensor is due to the presence of the active carbon filter. After 120 s, the output signal of each gas sensor reached 99% for the oxygenate

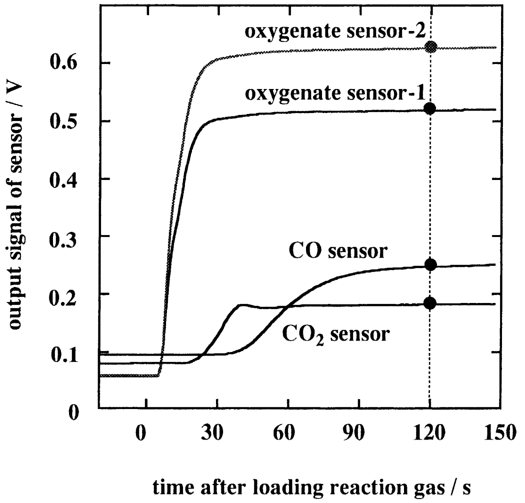


Fig. 6. Time courses of output signals of selective oxygenate sensors, CO sensor and CO_2 sensors after introduction of effluent.

sensors, 97% for the CO sensor and 100% for the CO_2 sensor. The response time of this gas sensor system is less than 2 min even for a transient change in the products. This is much faster than the time required for the precise gas analysis, 40–50 min, by a conventional FID–GC.

The effluent of the propane oxidation over $Cs/Fe/SiO_2$ (1/0.05/100) [18–20] at 400 to 500°C was analyzed by the gas sensor system and also the FID–GC equipped with a methanator. Fig. 7 shows the correlation between the concentration evaluated from the FID–GC and the output signals of the gas sensors. The output signals increased in proportion to the sum of the oxygenate yield on a logarithmic scale. The obtained fitting curve shown in Fig. 7 was used for the calculation of the sum of oxygenate yields on propane oxidation over alkali/Fe/SiO_2 (1/0.05/100).

Fig. 8 shows the product distributions determined by (a) a gas sensor system and (b) gas chromatography for propane oxidation over alkali/Fe/SiO_2 (1/0.05/100). Since little alcohol was produced, there was no large difference between the signals from

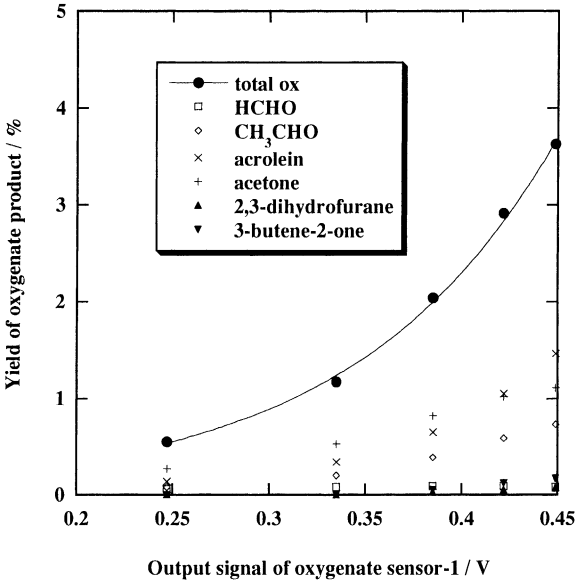


Fig. 7. Response of oxygenate selective sensor-1 toward the selective propane oxidation effluents over $Cs/Fe/SiO_2$ at 400, 425, 450, 475 and 500°C.

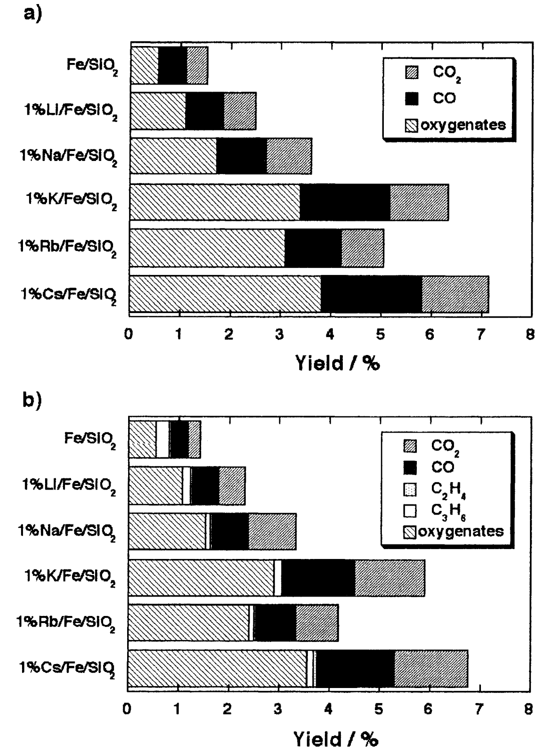


Fig. 8. Comparisons of the evaluation result by (a) the gas sensor system with that by (b) FID–GC for the propane oxidation effluents over Fe/SiO_2 and alkali modified Fe/SiO_2.

the oxygenate sensors-1 and -2. When we compared the oxygenate yield estimated from the gas sensor with that from the FID–GC, the largest difference was found in Rb/Fe/SiO_2 with 2.4% yield by the sensor and 3.0% yield by FID–GC. Although the difference is not small, the evaluation results obtained with gas sensor system is as same as those with FID–GC in the order of good catalysis. Both results indicated that the addition of potassium or cesium induces a high propane conversion and a high oxygenate yield. The gas sensor system has an advantage for the rapid evaluation of the selective oxidation catalysts.

The disadvantages of the oxygenate sensor are lower selectivity and the saturation of the output signal. The output signal of the oxygenate sensor increases in proportion to the logarithmic scale of the oxygenate concentration. At a higher concentration of oxygenate, the change in the output signal becomes smaller. This problem was solved by the dilution with air up to the optimal concentration for the oxygenate sensor. On the other hand, the distribution of the produced oxygenates with the same functional group cannot be analyzed by the oxygenate sensors employed here. As mentioned before, the low selectivity of gas sensors is an important issue. Chemometrics or pattern recognition of the signals from several different gas sensors will enable us to distinguish each gaseous compound.

4. Conclusions

Three types of catalytic oxidations, the simple combustion of CO, the oxidative dehydrogenation of ethane, and the oxygenate formation from propane were evaluated by the gas sensor systems. Although the accuracy of the gas analysis by the sensor system is lower than that of gas chromatography, the gas sensors have the advantages of a rapid response, continuous measurement, small size and are less expensive. Such small sized sensors will enable us to design miniaturized reactors in which the sensor is included. A two-dimensional (2-D) catalyst library deposited on a silicon wafer will be evaluated by the 2-D sensor arrays with a structure similar to the catalyst library.

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