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Environmental Gas Sensors

Duk-Dong Lee and Dae-Sik Lee

Abstract—Due to the dramatic growth in industrial development and population, the natural atmospheric environment has become polluted and is rapidly deteriorating. Thus, the monitoring and control of such pollutants is imperative to prevent environmental disasters. Conventional analytic instruments for this purpose are time consuming, expensive, and seldom used in real-time in the field. As such, a solid-state gas sensor that is compact, robust, with versatile applications and a low cost, could be an equally effective alternative. Accordingly, this paper presents a brief overview of solid-state gas sensors, which can be classified into semiconductor, capacitor, and solid-electrolyte type sensors, based on their sensing mechanisms and a simple NDIR instrument. Furthermore, the sensing properties of solid-state gas sensors to environmental gases, such as NO_x , SO_x , CO_2 , volatile organic compounds (VOCs), plus certain other gases, are also classified and summarized.

Index Terms—Gas sensor, odors, pollutants, NDIR, solid electrolyte, VOC.

I. INTRODUCTION

THE current era of high technology and advanced industry has produced an incredible rise in living standards. However, this has also been accompanied by a variety of serious environmental problems, for example, the release of various chemical pollutants, including NO_x , SO_x , HCl, CO_2 , volatile organic compounds (VOCs) and fluorocarbon, from industry, automobiles, and homes, into the atmosphere, resulting in global environmental issues, such as acid rain, the greenhouse effect, sick house syndrome, and ozone depletion.

Environmental pollution can be classified into three categories: atmospheric, water, and soil pollution. In particular, atmospheric pollution, along with water pollution, can cause major disasters within a short period of time, since this type of pollution can diffuse rapidly over large areas. Furthermore, since the kinds and quantities of pollution sources have also increased dramatically, the development of a method for monitoring and controlling these sources has become very important.

To prevent or minimize the damage caused by atmospheric pollution, monitoring and controlling systems are needed that can rapidly and reliably detect and quantify pollution sources within the range of the regulating standard values. Until now, air pollutant measurements have been carried out with analytic instruments using optical spectroscopy or gas chromatog-

TABLE I
COMPARISON BETWEEN ANALYTICAL INSTRUMENTS AND GAS SENSORS

	Analytical instruments (GC, UV)	Gas sensor
Resolution	Excellent	Comparable
Cost	Very high	Fair
Size	Bulky (Factory)	Compact
Rigidity	Fragile	Rigid (replaceable)
Process control	Difficult	Easy
Mass production	Difficult	Easy
Measurement	Instantaneous	Continuous

raphy/mass spectrometry. However, although these instruments can give a precise analysis, they are time-consuming, expensive, and can seldom be used in real-time in the field. As such, a gas sensor that is compact, robust, with versatile applications and a low cost, could be an equally effective alternative. Table I presents several feature comparisons between gas sensors and analytical instruments. A lot of recent research and development has been focused on the development of solid-state gas sensors, consequently, their performances have improved dramatically.

There are several solid-state gas sensors currently available for gases such as O_2 , H_2O , and LNG at relatively high concentrations. However, the range of air pollutant concentrations that can be detected only reaches as low as ppm (10^{-6}) in combustion exhaust control or indoor monitoring and ppb (10^{-9}) in atmospheric environmental monitoring. Therefore, the development of more sensitive and selective gas sensors than the above conventional sensors is still required.

This paper briefly introduces several gas sensors that have been developed for environmental monitoring along with their sensing principles.

II. ATMOSPHERIC POLLUTION AND EMITTING SOURCES

A. Atmospheric Pollution

Atmospheric pollution is defined as a status containing gases, offensive odors, and particles that are harmful to humans, animals, vegetables, or living environments above the regulation limits in specific regions. Atmospheric pollution has become particularly serious since the Industrial Revolution. Furthermore, due to the recent dramatic growth in population and industrial development, along with an intensified usage of fossil fuels, the natural atmospheric environment has become polluted and is rapidly deteriorating. Yet, since the level of public concern related to living and working in a healthy environment has now increased, the demand for monitoring and controlling the atmospheric environment in the house and

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D.-D. Lee is with the School of Electronic and Electrical Engineering, Kyungpook National University, Taegu, Korea (e-mail: ddlee@ee.knu.ac.kr).

D.-S. Lee is with the Microsystem team, Telecommunications Basic Research Laboratory, Electronics and Telecommunications Research Institute (ETRI), Taegon, Korea (e-mail: dslee@etri.re.kr).

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TABLE II
AIR POLLUTANTS, THEIR REGULATION VALUES, AND DETECTION METHODS

Gas	Regulation values (ppm)			Detection method
	In air	In exhaust	In workplace (TLV-TWA ^(a))	
CO ₂	350-500	10 ⁴ -10 ⁵	5000	IR absorption Ion-selective electrode
NO _x	10 ⁻² -10	1-1000	3-25	IR absorption Flame emission spectrometry Mass Spectrometry
SO ₂	10 ⁻² -10	10-1000	3	Flame Emission Spectrometry UV absorption
O ₃	10 ⁻³ -1		0.1	UV absorption
Hydrogen Chloride (HCl)		1-50	5	IR absorption Flame Emission Spectrometry Mass Spectrometry
Benzene (C ₆ H ₆)		1-50	10	Gas chromatography Mass spectroscopy
CO	1-50	10-1000	50	IR absorption Flame Emission Spectrometry Mass Spectrometry Electrochemical Sensors

workplace has also increased. As a result, intensive research efforts have been made in various fields in an attempt to resolve such environmental problems.

B. Pollution-Emitting Sources and Regulating Standards

Emitting sources can be classified into two categories according to the state of the source, that is, a stationary or mobile source. Stationary sources include houses, workplaces, thermal power stations, and so on, whereas mobile sources include automobiles, trains, ships, and so forth. Stationary emitting sources also cover many diverse fields, such as oil refineries, chemical plants, metal refineries, and grocery plants. In particular, with the tremendous increase in automobiles and consumption of fossil fuel, the atmospheric environment has deteriorated significantly. Therefore, the regulation of pollutant emissions has intensified all over the world. Table II shows some atmospheric pollution molecules and several environmental standard values.

III. SENSING PRINCIPLES OF SOLID-STATE GAS SENSORS

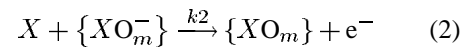
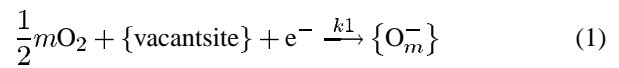
Gas sensors for detecting air pollutants must be able to operate stably under deleterious conditions, including chemical and/or thermal attack. Therefore, solid-state gas sensors would appear to be the most appropriate in terms of their practical robustness. Even though there are many kinds of solid-state gas sensors, as shown in Table III, only solid electrolyte, semiconductor, and nondispersive infrared absorption types are normally utilized. Four representative sensors are briefly described here.

A. Semiconductor and Capacitor-Type Gas Sensors

Semiconductor gas sensors are widely used for detecting inflammable gases and certain toxic gases in air. The adsorption or reaction of a gas on the surface of the semi-conducting material induces a change in the density of the conducting electrons in the polycrystalline sensor element. This chemical reaction can be described by four steps as follows.

- 1) Pre-adsorption of oxygen on semi-conducting material surface.
- 2) Adsorption of specific gas.
- 3) Reaction between oxygen and adsorbed gas.
- 4) Desorption of reacted gas on surface.

The above process of delivering electrons between the gas and the semiconductor actually represents the gas sensitivity of the gas sensor. When reacted with the gas, the conductivity of the semiconductor gas sensors increases when the adsorbed oxygen molecules play the role of acceptor, whereas the conductivity decreases when the adsorbed oxygen molecules play the role of donor. The principle is based upon initial reversible reaction of atmospheric oxygen with lattice vacancies in the oxide and the concurrent reduction in electron concentration n . This reaction generates various oxygen species, (denoted by the constant m), according to the temperature and oxygen pressures, i.e., O₂⁻, O⁻ or O²⁻, which can then react irreversibly with certain combustible species. As such, the basic reactions occurring at the sensor surface can be represented as follows [46]



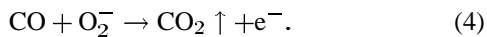
where the species in curly braces are bound on surface sites, m is integer and X is a combustible species, such as methane. As shown by (1)–(3), the concentration of oxygen species present at the surface of the tin oxide particles will depend on the balance between the rates of the first two reactions, with increasing the concentrations of the combustible species, X , the steady-state concentration of O_{*m*}⁻ will decrease. From (1)–(3), it is clear that the presence of oxygen is essential for the operation of metal oxide compound semiconductor sensors.

These oxygen molecules are preadsorbed on the semiconductor surface, then when they react with specific gases like CO and hydrocarbons, this produces changes in the electrical con-

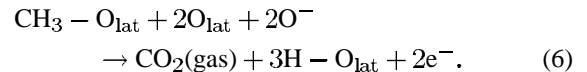
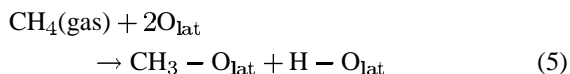
TABLE III
SOLID-STATE GAS SENSORS FOR DETECTING ENVIRONMENTAL GASES

Types of sensor and sensing materials	Typical detection ranges	Operating temp. (°C)	Year	Ref.
NO₂				
Semiconductor type				
Thick film PdAu-SnO ₂	50 ppm	250	1885	1
Thick film In ₂ O ₃	10-100 ppm	250	1989	2
Thick film WO ₃	20-80 ppm	300	1991	3
PbPc	0.04-10 ppm	150	1989	4
Thin film Cd-SnO ₂	10-100 ppm	300	1991	5
Thin film PdPtAu-WO ₃	1-10 ppm	200	1998	6
Thick film TiO ₂ -WO ₃	0.5-50 ppm	350	2000	7
WO ₃ micro sensor	1-50 ppm	300	2000	8
Solid-electrolyte type				
Ba(NO ₃) ₂	5-800 ppm	480	1977	9
Na-β/β''-alumina/NaNO ₃	20-1%	160	1987	10
Na-β/β''-alumina/Ba(NO ₃) ₂	10-200 ppm	450	1991	11
NASICON/NaNO ₂ +Li ₂ CO ₃	0.005-200 ppm	150	1993	12
Y ₂ O ₃ -ZrO ₂ /CdCr ₂ O ₄	20-200 ppm	500	1997	13
NASICON/pyrochlore oxide	10-1000 ppm	400	2000	14
Capacitor type				
NiO/ZnO	10-100 ppm	300	1995	15
NO				
Semiconductor type				
Thin film In ₂ O ₃ -SnO ₂	2-1000 ppm	200	1988	16
Thick film WO ₃	20-80 ppm	300		3,17
Thick film Bi ₂ O ₃ -WO ₃	2-300 ppm	300	1998	18
Thin film PdPtAu-WO ₃	1-10 ppm	200	1998	6
Solid-electrolyte type				
NASICON/NaNO ₂	1-800 ppm	150	1992	19
Y ₂ O ₃ -ZrO ₂ /CdCr ₂ O ₄	20-200 ppm	500	1997	13
Capacitor type				
SrSnO ₃ -WO ₃	1-400 ppm	550	2000	20
SO_x				
Semiconductor type				
Ceramic SnO ₂	20-200 ppm	220	1984	21
Solid-electrolyte type				
K ₂ SO ₄	10 ppm – 1%	820	1977	9

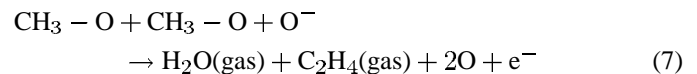
ductivity due to an oxidizing reaction. For example, when CO reacts with the preadsorbed oxygen molecules, CO₂ gas is generated as follows [47]



As another example, CH₄ is thought to react with oxygen molecules on the surface as follows [48]



Simultaneously



where O_{lat} is lattice oxygen in the metal oxide semiconductor.

The theory predicts that increasing the concentration of a combustible gas will lead to an increase in carrier concentration, e⁻ in (2), (4), (6), and (7). If we denote the concentration

TABLE III (Continued.)
SOLID-STATE GAS SENSORS FOR DETECTING ENVIRONMENTAL GASES

Types of sensor and sensing materials	Typical detection ranges	Operating temp. (°C)	Year	Ref.
NASICON/Na ₂ SO ₄	50 ppm – 1%	780	1985	22
Na-β/β''-alumina/Na ₂ SO ₄	40 ppm - 20 %	500 - 900	1988	23
MgO-ZrO ₂ /Li ₂ SO ₄ -CaSO ₄	20-200 ppm	700	1992	24
NASICON/Na ₂ SO ₄ ,BaSO ₄	5 – 100 ppm	300 - 550	1996	25
Y ₂ O ₃ -ZrO ₂ /BaSO ₄ ,K ₂ SO ₄ ,SiO ₂	20-10000 ppm	650 – 1000	2000	26
CO₂				
Semiconductor type				
La ₂ O ₃ -SnO ₂	100-3000	400	1991	27
Solid-electrolyte type				
K ₂ CO ₃	50 - 1000 ppm	450-650	1977	9
NASICON/Na ₂ CO ₃	50 – 1000 ppm	530	1984	28
NASICON/Na ₂ CO ₃ -BaCO ₃	4 ppm – 40 %	550	1990	29
NASICON/Li ₂ CO ₃ -CaCO ₃	100 ppm – 20%	500	1991	30
NASICON/NdCoO ₃	100 – 2000 ppm	200-300	2000	31
LISICON/Li ₂ CO ₃ ,K ₂ CO ₃ ,Na ₂ CO ₃	500 – 10000 ppm	420	2000	32
Capacitor type				
CuO-BaTiO ₃	50ppm – 10%	450-600	1995	33
AMO/PTMS	100-3000	60	1999	34
CeO ₂ /BaCO ₃ /CuO	350 - 20000	550 - 650	2000	35
O₃				
Semiconductor				
Thin film SiO ₂ -CeO ₂ -In ₂ O ₃	0.05-5 ppm	420	1987	36, 37
Thick film Fe ₂ O ₃ -In ₂ O ₃	0.01-5 ppm	350	1992	38
Thin film WO ₃	10 – 160 ppb	200-400	2000	39
Thin film Zn ₂ In ₂ O ₅ -MgIn ₂ O ₄	0.4 – 6 ppm	275	2000	40
VOCs				
Thin film Pt-SnO ₂	1-1000 ppm	250-300	1996	41
Thin film Al,Pt,Pd-SnO ₂	50-5000 ppm	300	1997	42
Polymer (Polypyrrole)	0 – 15 % (methanol)	25	1989	43
Polymer (polypyrrole)	1 – 1000 ppm	20	1996	44
Polymer (polyaniline)	1 – 350 ppm	20	1996	45

of carriers as n , then we can write the following expression for the electrical conductivity of the material [47]:

$$\Delta\sigma = q\mu_n n \propto [X]^r; \text{ where } 0.5 < r > 1 \quad (8)$$

where

- μ_n mobility of the carrier (i.e., electron);
- q electron charge;
- X combustible species, such as methane.

Although the modulation in carrier concentration can strongly affect the surface conductance in thin single-crystal films, it is unable to explain the large effect observed in thick and ceramic metal oxide films. Therefore, the model of conduction must be extended to consider a concentration-dependent mobility term that arises from polycrystalline oxides having an open granular structure into which the gas can diffuse and react at the surfaces of the granules. The electron mobility μ_n is then governed by the potential barrier between neighboring grains in a hopping model.

C. Xu and N. Yamazoe [49] reported that nano-scale minute particles under 6 nm show a tremendous increase in sensitivity because of the relative increase in the ratio of the surface area to

the bulk. To explain the sensing-mechanisms for inter-granular contacts, the neck model can be used for SnO₂ with nano-scale grains smaller than 6 nm in diameter, whereas both the neck and the grain boundary models can be used for grains larger than 6 nm.

Therefore, any change in conductivity ($\Delta\sigma$) is mainly related to the specific gas concentration through its effect on the electron concentration (n) and electron mobility (μ_n). This change is modified according to the kind of additives, grain sizes, contacts and the states of surface and bulk [50].

In addition, the electrical conductivity (σ) in a semiconductor sensor is also related to the oxygen partial pressure (P_{O_2}) [47]

$$\sigma \cong P_{O_2}^{-m} \quad (9)$$

where the exponent m lies between 0.5 and 1.0 according to the kind of reaction, such as the pore density or microstructure on the surface. In ZnO, TiO₂, or SnO₂, all widely-used materials, the exponent m is known to be about 0.5. Both the ambient oxygen partial pressure and the humidity could be disadvantageous in a practical viewpoint because they affect the sensitivity of metal oxide gas sensors.

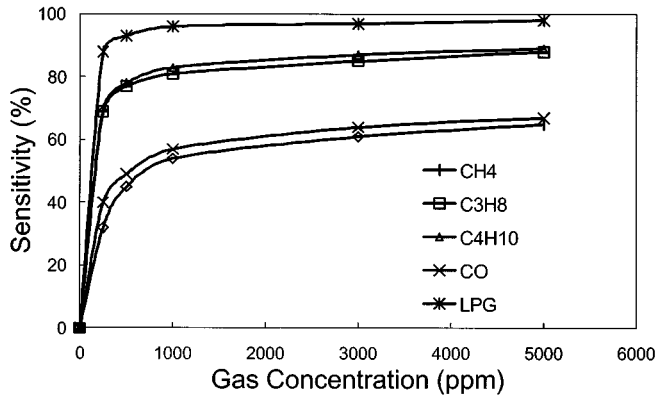


Fig. 1. Sensitivity of SnO_2/Ca , Pt gas sensor to combustible gases at 350°C .

Even though the sensitivity, i.e., the resistance variation of a sensor, is an important parameter, three other parameters, selectivity, stability, and speed, are also vital parameters. While it is difficult to obtain a reliable sensor that includes all these qualities, they have been relatively enhanced by controlling the material, structure, and operating conditions of the sensor. For example, the sensitivity of a SnO_2/Ca , Pt sensor, as a typical solid-state sensor, to several combustible gases, is shown in Fig. 1, at 350°C . The SnO_2/Ca , Pt sensor exhibits the highest sensitivity to butane and a good linearity to combustible gases. Different sensitivity patterns, even with the same sensor, have been found when modulating the operating temperature, and this has been utilized to recognize different kinds of gases in an electronic nose.

In case of capacitor-type gas sensors, they can measure the change of the dielectric constant of films between the electrodes as a function of gas concentrations. For an example, a spin-coated polyphenylacetylene conducting polymer film responds to the various gases, such as CH_4 , N_2 , CO , and CO_2 [56]. Capacitance changes in the capacitor-type sensors are typically in the range of pF and very dependent on the operating frequency and surrounding conditions, like humidity and temperature. Recently, a capacitive microsystem with the ability to measure at different sensor temperatures for discrimination of ethanol, toluene and their mixture, was also introduced [57].

B. Non-Dispersive Infrared Method Using Pyroelectric Infrared Sensors

The use of infrared for atmospheric environmental measuring, especially for monitoring exhaust gas, is focused on selective and reliable gas detection. It is generally recognized that nondispersive methods are simpler in structure and easier to use than dispersive methods.

Gas molecules selectively absorb the energy that corresponds to their own quantized vibrating energy, which is usually located in an infrared region. For example, CO_2 , CO , and CH_4 have a unique absorbing spectrum at 4.25 , 4.7 , and $3.3\ \mu\text{m}$, respectively, in an infrared region. The gas-absorbing degree is proportional and follows the gas concentration to be detected.

The absorptivity $A(\lambda)$ at a specific wavelength [58] can be defined as

$$A(\lambda) = E(\lambda)bc \quad (10)$$

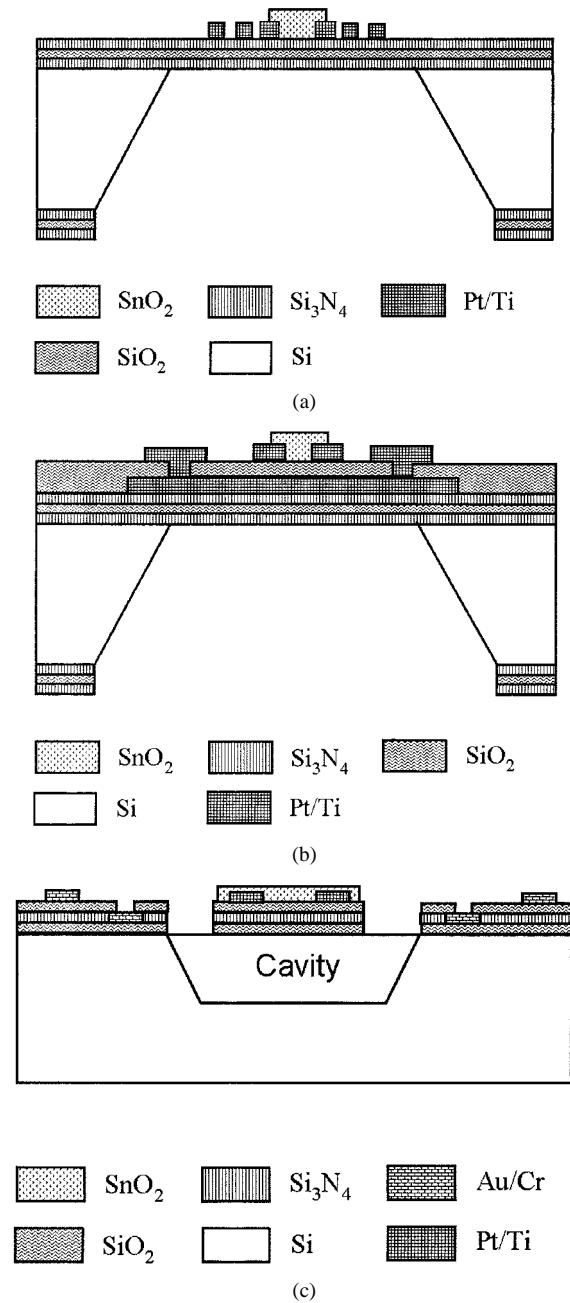


Fig. 2. Cross section of silicon-based micro gas sensor for low power consumption with (a) lateral heater, (b) vertical (embedded) heater, and (c) suspended heater.

$$A(\lambda) = -\log \left[\frac{I(\lambda)}{I_0(\lambda)} \right] \quad (11)$$

where

- $E(\lambda)$ proportionality constant called the absorptivity;
- b path length;
- c absorbing gas concentration;
- $I_0(\lambda)$ incident beam power.

As shown in these equations, the absorbance is directly proportional to the path length through a gas and the concentration of the absorbing species. Therefore, by using a monochromatic beam with a large absorptivity $E(\lambda)$ to measure the intensity of a beam after it passes through a gas, the concentration of the gas to be detected can be determined without interfering with the other

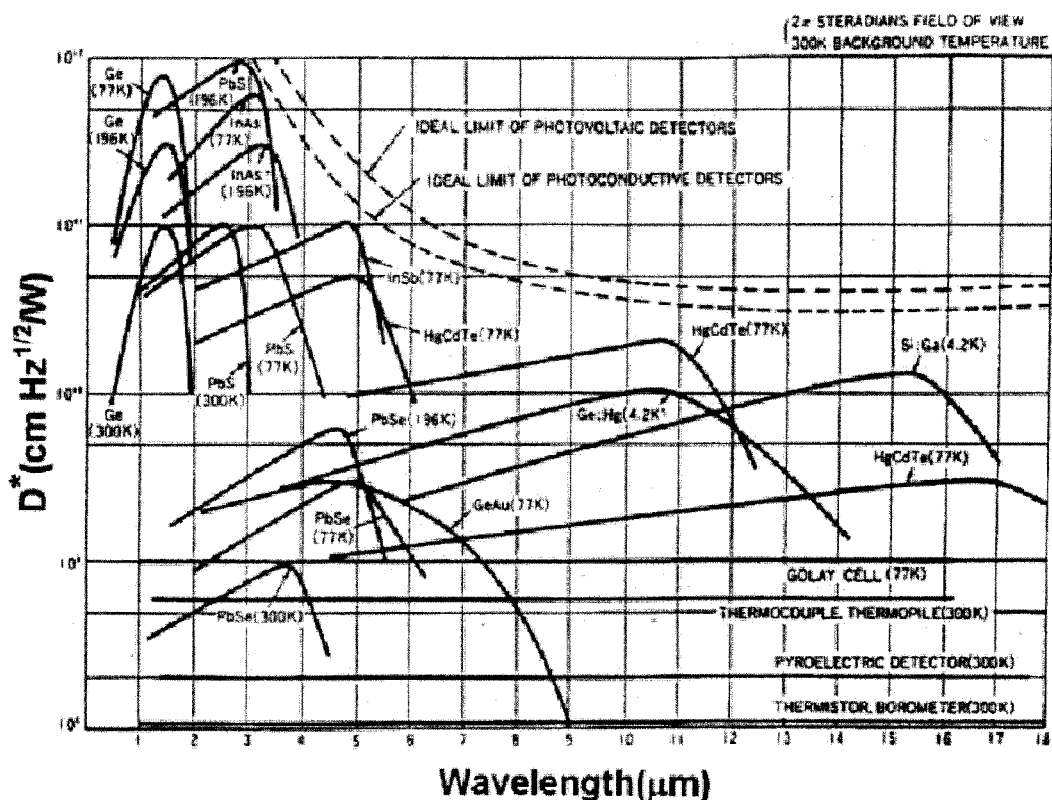


Fig. 3. Construction materials for infrared detector.

gases. For most spectroscopic analyzes, radiation consisting of a limited, narrow, and continuous group of wavelengths, called a band, is required. A narrow bandwidth tends to enhance the sensitivity of the absorbance measurements and can provide selectivity to both emission and absorption methods. In the NDIR gas-detecting method, filters or monochromators are used to obtain a monochromatic beam from an infrared light source with a wide wavelength range.

Fig. 3 shows the spectral responses from different infrared light detectors with upper-limiting temperatures [59].

With a wavelength range of $4 \sim 5 \mu\text{m}$, InSb, PbSe, and HgCdTe have been used as the sensing materials for an infrared detector. However, they need to be used within a low temperature range of $77 \text{ K} \sim 196 \text{ K}$. InAs, PbS, and PbSe have also been used as the sensing materials for an infrared detector, within a wavelength range of $3 \sim 5 \mu\text{m}$ for measuring HC gases. Recently, IR sensors for detecting environmental gases, like CO_2 , CO, and HC, along with a chromatic filter, have been used in the NDIR method. The use of a compensating circuit for thermal noise elimination is also needed in this case. Fig. 4 shows a hardware block diagram for an environmental monitoring system using the nondispersive infrared method. The output voltage in an IR sensor is directly proportional to the gas concentration because the absorptivity of the incident beam, when the gas passes through the cell, is proportional to the path length and the gas concentration, according to the Beer-Lambert law [60]. To gain a high sensitivity, it is necessary to keep the cell length appropriately long. For example, a 20 cm-long cell is sufficient to achieve a reliable sensitivity in the case of monitoring indoor gases. Fig. 5 shows the output signals in the case of monitoring

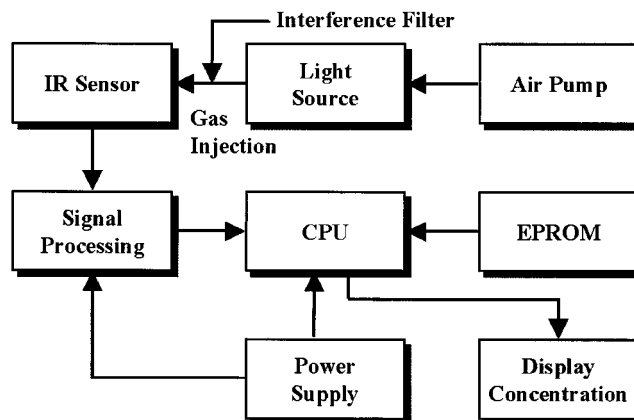


Fig. 4. Hardware block diagram for NDIR-type gas instrument.

CO_2 gas repetitively, indicating a good sensitivity and reproducibility.

C. Solid-Electrolyte Gas Sensors

Since a NASICON solid-electrolyte potentiometric gas sensor using alkali metal carbonate as an auxiliary phase solid electrolyte is known to be sensitive to CO_2 , a lot of recent research has focused on the development of compact NASICON sensors. This type of solid electrolyte sensor can be used for the detection of NO_x or SO_x . Three types have been classified by Weppner [61], based on whether the ionic species derived from the gas to be detected coincides with the mobile ion (Type I), the immobile ion (Type II), or neither of them (Type III) of the solid electrolyte used, as shown in Fig. 6. With

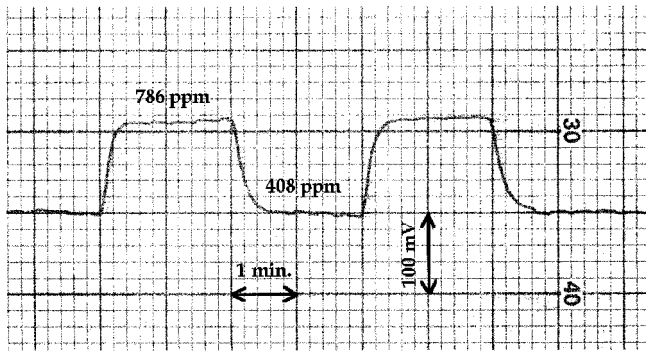


Fig. 5. Time response of NDIR-type CO₂ gas instrument.

a Type III sensor, an auxiliary phase attached to the surface of the solid electrolyte is needed. For this auxiliary phase, a compound is used that contains the same ionic species as those from the gas. The auxiliary phase acts as a poor ion-conducting solid electrolyte which, by attaching to the sensing electrode, forms half of a Type I or II. This combination then makes up the electrochemical cell, as shown in Fig. 6. Type III sensors can be divided into several subgroups depending on the type of the half-cells combined. Type III sensors are used for detecting oxygenic gases, such as CO₂, NO_x and SO_x.

There is also a LISICON gas sensor that uses Li⁺ instead of Na⁺ as the ionic conductor. This sensor exhibits a high sensitivity to CO₂, even at a lower device temperature. Fig. 7 shows the structure of a LISICON sensor for detecting CO₂, which demonstrates a good linearity to CO₂ gas along with a high stability through the use of inorganic binders for the auxiliary phase.

IV. SENSING OF ENVIRONMENTAL GASES

A. NO_x Sensing

NO_x (NO and NO₂) gas is known to be very harmful to humans and one of the main causes of acid rain. Solid-state sensors for NO_x are desperately needed for monitoring the combustible exhaust in the environmental atmosphere. Only a few sensors are currently commercially available. As NO and NO₂ have quite different properties from each other, there are hardly any reports on gas sensors that respond to both NO and NO₂ with the same sensitivity. Most research has focused on the development of NO₂ sensors. Among the various existing NO₂ sensors, semiconducting oxides and solid electrolytes would appear to be the best.

Semiconductor NO₂ sensors using metal oxides have been investigated by many researchers. The sensitivity to NO₂ and NO is dependent on the oxide used. Until now, WO₃-based sensors have shown an excellent sensitivity to NO₂ above 1 ppm in air at an operating temperature range of 250–350 °C, as shown in Fig. 8, where the sensitivity is defined as the ratio of the electrical resistance of the sensor in the gas (R_g) to that in air (R_a). The same device also shows a high sensitivity to NO, although this sensitivity is much lower than that for NO₂. Therefore, to be applicable for controlling combustion exhaust, the NO sensitivity needs to be improved. The addition of noble metals (Pt,

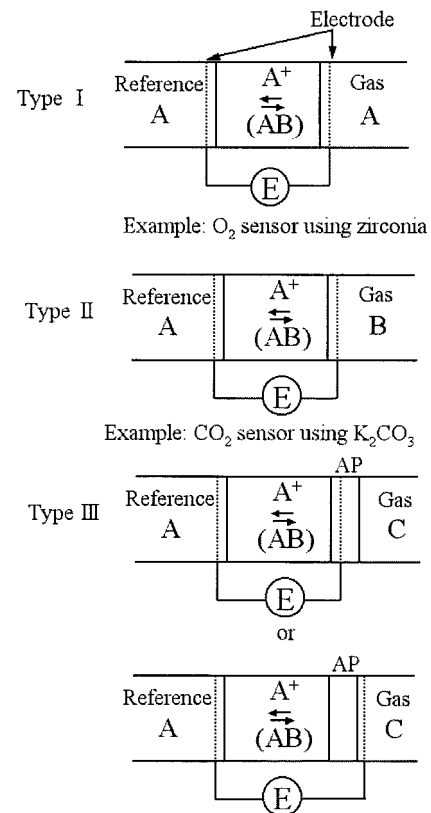


Fig. 6. Three types of solid-electrolyte gas sensor.

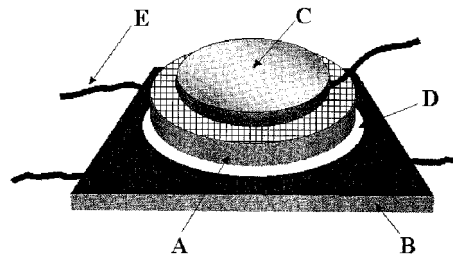


Fig. 7. Structure of LISICON CO₂ gas sensor; (1) Li⁺ ionic conductor, (2) Al₂O₃, (3) alkali metal carbonate, and (4) inorganic encapsulant.

Ru and Au) [17] or metal oxides (TiO₂ and SnO₂) to the WO₃ material could enhance the sensitivity [7].

Solid-electrolyte NO₂ sensors can be fabricated using an Na⁺ conductor (solid electrolyte) and auxiliary phase, like NaNO₃. The e.m.f. of such a sensor can be expressed by

$$E = E_0 + \frac{RT}{F} \ln P_{\text{NO}_2} \quad (12)$$

regardless of the presence of coexistent oxygen. With the proper selection of the auxiliary phase, the NO₂-sensitivity can be improved to lower the detecting limit to less than 0.2 ppm. As for NO, more research is still required because NO sensing is more important than NO₂ sensing for combustion exhaust control.

B. SO_x Sensing

SO_x (SO₂ and SO₃) gas in the global atmosphere is a major source of acid rain. The major industrial sources

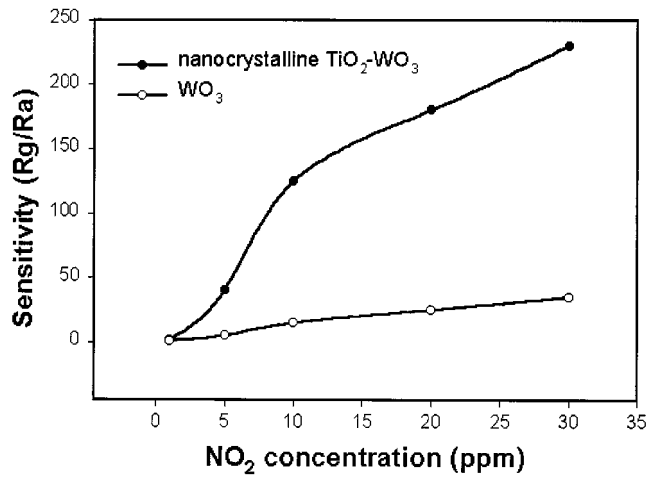
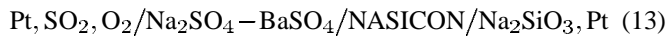


Fig. 8. Sensitivity of $\text{TiO}_2\text{-WO}_3$ and WO_3 gas sensors as a function of NO_2 gas concentration.

of SO_2 emissions are coal-fired power plants, oil and gas productions, and nonferrous smelting. Currently, chemical analyses (West-Gaeke coulometric technique, hydrogen peroxide method) and instrumental analyses (flame photometric detection, UV fluorescence technique) are used to determine SO_2 gas in stack gases, however, these methods are very complicated. Therefore, great efforts have been made to find more suitable methods for the continuous monitoring of SO_2 gases. One proposal is to use solid-electrolyte-type devices. Various sulfate-based solid electrolytes have been applied to Type III sensors, thereby combining NASICON or another Na^+ conductor (solid electrolyte) and Na_2SO_4 (auxiliary phase). These sensors have shown a high sensitivity and linearity to SO_x gas concentrations. For example, the structure of a NASICON SO_2 sensor can be shown as follows [25]



where Na_2SiO_3 is the reference electrode in air and the binary composite of Na_2SO_4 and BaSO_4 is the auxiliary phase. The e.m.f. of these sensors can be expressed as follows for the detection of SO_x in air

$$E = E_0 + \frac{RT}{2F} \ln P_{\text{SO}_x} \quad (14)$$

The e.m.f. of an NASICON SO_x sensor with an Na_2SiO_3 reference electrode and Na_2SO_4 auxiliary electrolyte is proportional to the SO_2 gas concentration, as shown in Fig. 9 [25].

A semi-conducting SO_2 sensor using ceramic SnO_2 was reported on for the first time by R.Lalauze *et al.* [21].

C. CO_2 Sensing

CO_2 is the main cause of the greenhouse effect, yet, by itself, it is harmless. CO_2 sensing is necessary for the auto-ventilation of air in living rooms and automobiles as well as for measuring or controlling bio-related activities.

A nondispersive infrared method is currently prevalent for monitoring CO_2 gas. Generally, infrared spectral regions can be classified as a near region ($0.8 \sim 2.5 \mu\text{m}$), middle region

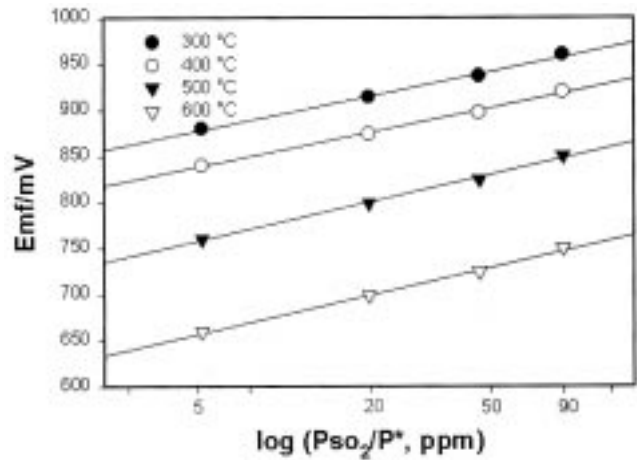


Fig. 9. EMF response of NASICON SO_x sensor with Na_2SiO_3 reference electrode and Na_2SO_4 auxiliary electrolyte to SO_2 gas.

($2.5 \sim 50 \mu\text{m}$), or far region ($50 \sim 1000 \mu\text{m}$). The region used the most is ($2.5 \sim 15 \mu\text{m}$). For CO_2 , the main absorption wavelength is known to be $4.25 \mu\text{m}$. By using a filter that only transmits a beam with a $4.25 \mu\text{m}$ -wavelength, the absorbance beam can be detected by an infrared beam detector. The output can then be expressed as a function of the CO_2 gas concentration. For example, the response of a CO_2 NDIR sensor is shown in Fig. 5, where the gas concentration was changed from 400 to 800 ppm.

Since the first solid-state CO_2 sensor based on electrochemical principles (Type II) was reported in 1977 by Gauthier and Chamberland [62], a lot more research has been conducted. Nowadays, NASICON ($\text{Na}_3\text{Zr}_2\text{Si}_2\text{PO}_{12}$) or LISICON ($\text{Li}_3\text{Zr}_2\text{Si}_2\text{PO}_{12}$) CO_2 sensors are used with an attached auxiliary phase of an alkali metal carbonate, such as Na_2CO_3 or Li_2CO_3 . A planar-type LISICON sensor element is shown in Fig. 7. In this structure, the potential at the sensing electrode contacting the auxiliary phase changes as a function of the partial pressure of CO_2 (P_{CO_2}), while the potential at the opposite electrode, set by the partial pressure of oxygen (P_{O_2}), is determined at a level matching the P_{O_2} in the air. The electromotive force of this sensor can be expressed by a Nernstian equation as follows

$$E = E_0 + \frac{RT}{2F} \ln P_{\text{CO}_2} \quad (15)$$

where

- E_0 constant;
- F Faraday constant;
- R gas constant;
- T temperature.

The Nernstian correlation holds very well over a wide range of CO_2 concentrations. The auxiliary phase is very important for Type III CO_2 sensors and multiple carbonate systems are known to enhance the stability of these sensors, for example, in relation to humidity. NASICON or LISICON sensors are recognized as exhibiting a good linearity to CO_2 gas concentrations. Fig. 10 shows the time response of LISICON CO_2 sensors and indicates fast reactions to the gas concentrations [32].

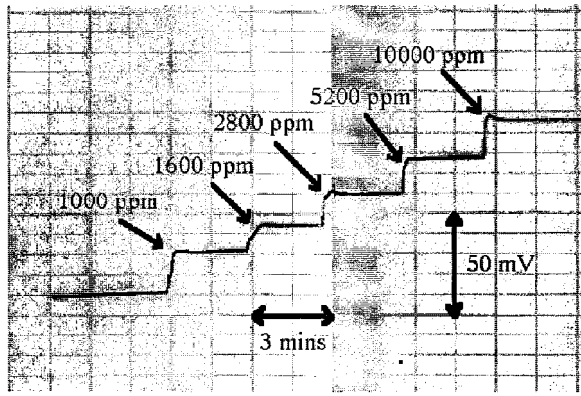


Fig. 10. Time response of LISICON CO₂ gas sensor.

Capacitor-type CO₂ sensors are implemented using composite ceramics, like CeO-BaCO₃/CuO [63]. These sensors exhibit a high sensitivity to CO₂ plus selectivity within a concentration range of 100 ~ 100 000 ppm. Furthermore, a semi-conducting CO₂ sensor using La₂O₃ - SnO₂ was recently reported [64].

D. Sensing of Other Gases

Solid-state gas sensors for detecting small concentrations of other pollutants, such as ozone [65], [66], HCl [67], [68], hydrocarbons [69]–[71], carbon monoxide [72]–[74], H₂S [75]–[77], NH₃ [78], [79], volatile organic compounds (VOCs) etc., in air, are also important for environmental conservation and a comfortable surrounding for humans. As for ozone, the detection of several ppm concentrations in air has been reported with an In₂O₃-based element, thereby making it possible to monitor the environmental ozone [80]–[82]. Due to the severe regulations on the emission of VOCs, known as the main cause of sick-house syndrome, there has been a lot of recent research on VOC-sensing materials, such as metal oxides, polymers and quartz crystal microbalances (QCM), and the resulting sensitivity and stability has indicated a good ability to detect environmental VOCs [83]–[89]. Further research and development on solid-state sensors for other miscellaneous pollutants is still needed.

V. CONCLUSION

This paper presented a brief overview of atmospheric pollution and emitting sources, the sensing mechanisms of solid-state sensors, and the sensing of gases in sensors. When considering the hazards of environmental pollution that can cause severe injury within a short time period, the precise monitoring of pollutant emissions is very important to prevent environmental accidents. Conventional instruments for monitoring the environment are not so popular because they are bulky, time consuming, and expensive. Accordingly, compact, robust, and inexpensive solid-state gas sensors are required as an effective alternative for environmental monitoring. In addition, the recent escalation in the desire to live in a comfortable environment along with significant advances in sensor technologies for monitoring pollutant gases have resulted in a more focused and rapid development of environmental gas sensors.

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Duk-Dong Lee was born in Taegu, Korea, in 1942. He received the B.S. degree in physics and the M.E. degree in electronics from Kyungpook National University, Taegu, Seoul, Korea, in 1966 and 1974, respectively, and the Ph. D. degree from Yonsei University, Seoul, Korea, in 1984.

He is currently a professor at the School of Electronic and Electrical Engineering, Kyungpook National University. He has performed research on semiconductor gas sensors since 1978 and also researches in the field of thin and thick film gas sensor, the NDIR gas-detecting system, and electronic nose system.

Dae-Sik Lee was born in Kyungju, Korea, in 1970. He received the B. Eng. degree in 1995 and the M. Eng. degree in 1997 from Kyungpook National University, Taegu, Korea. He received the Ph. D. degree from the Department of Electronic Engineering, Kyungpook National University, in 2000.

At present, he is a Senior Researcher at Basic Research Laboratory, Electronics and Telecommunication Research Institute (ETRI), Taegon, Korea. His current research interests are MEMS, gas sensors, and electronic nose system.