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Malodor Detection Based on Electronic Nose

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1. Introduction

Recent decades have observed significantly increasing interest in the applications of electronic nose (E-nose) for qualitative analysis of odors. The first E-nose experiments were conducted in the early 1990s. (Shurmer et al., 1990; Shurmer & Gardner, 1992) Since then, E-nose has become a powerful tool to complement or even replace traditional chemical analysis in many applications ranging from quality control of foods (Barié et al., 2006; Panigrahi et al., 2006; Santonico et al., 2008; Tikk et al., 2008) and beverages (Ragazzo-Sanchez et al., 2008; Yu & Wang, 2007; Wongchoosuk et al., 2009b, 2010b), environment protection (Negri & Reich, 2001; Kuske et al., 2005), medical applications (Chan et al.,2009) to public safety (Scorsone et al., 2006; Zhang et al., 2007). Electronic nose employs an array of chemical gas sensors, numbering from 2 up to a few hundred sensors. Research on chemical gas sensors is mainly focused on improving two properties: selectivity (specificity to a molecule or a class of molecules) and sensitivity (strength of signal upon exposure to low concentration of molecules). Both selectivity and sensitivity leads to performance enhancement of an e-nose for specific applications such as bomb detection (Lubczyk et al., 2010), determination of food freshness based on amine detection (Lorwongtragool et al., 2011; Liao et al., 2010), quality control of alcoholic beverages (Wongchoosuk et al., 2009b, 2010b) and hydrogen gas sensing (Wongchoosuk et al., 2010a).

Chemical sensors can be classified into 4 types (James et al., 2005) based on their transduction, a mechanism that converts chemical interaction into a sensor signal: (1) Optical, (2) Thermal, (3) Electrochemical and (4) Gravimetric. Electrochemical transduction has so far dominated research activities on gas sensors, because its interface setup is more straightforward than other transduction methods. (Choopun et al., 2007; Lorwongtragool et al., 2011) Nevertheless, many research groups including us have been working on many transduction principles, i.e., optical (Uttiya et al, 2008) and gravimetric (Tuantranont et al, 2008), in parallel in order to take advantage of hybrid methodology that could dramatically enhance the performance of electronic nose. Due to the simplicity of the electronics involved, most commercial chemical gas sensors adopt electrical transduction technology in which the metal oxide semiconductors assume the most used sensor architecture according to their low-cost, high sensitivity and simplicity in function. (Korotcenkov, 2007) Thus, one

could easily integrate several functional elements such as sensitive layer, signal converter and control electronics within the same device of a size as required for most applications. Notwithstanding the simple working principles of metal oxide gas sensors, the gas-sensing mechanism at the microscopic level is quite complex and so far still insufficiently understood. (Korotcenkov, 2005; Surnev et al., 2003) As widely known, the gas sensors having the same metal oxide materials can have completely different gas-sensing properties depending on the preparation conditions. It is commonly believed that the chemo-resistive change of the metal oxides is caused by catalytic redox reactions at the sensing surface. Such reactions are controlled by electronic structure, chemical composition, crystal structure and relative orientation of the oxide surface to the analyte molecules, thereby allowing tuning their gas-sensing properties by modifying such parameters. Structural engineering by reducing grain size and modifying crystallite microstructure has been widely accepted as the best method to optimize the metal oxide gas sensors. It can be said that metal oxide gas sensors have nowadays been the most robust technology for gas detection in most applications. As will be seen in this chapter, most E-noses employ this type of gas sensors, except for section 2.1 where carbon nanotube/polymer gas sensors were developed for amine detection.

In this chapter, we have explored the applications of E-nose for malodor detection in three areas: (1) animal malodor, (2) human body odor and (3) indoor air quality.

2. Animal malodor

Among the numerous applications of an E-nose, detection of malodor involving with animal often refers to odor assessments of animal products and animal waste. The changes in the sensory properties of the both animal products and waste are mainly caused by the decomposition of organic matters. The major cause of these unpleasant and unacceptable odors is the growth and metabolism of microorganisms such as bacteria, yeasts and moulds leading to formation of the various toxic volatile organic compounds (VOCs) (Gram & Dalgaard, 2002; Huis in't Veld, 1996). In this section, a review of E-nose applications for 54malodor detection as caused by animal products and animal waste is given.

2.1 Animal products

Quality of animal products is very important because they are protein sources for human and some animals. Mechanisms of food spoilage can be classified into four types, as follows (Berk, 2009; Ghaly et al., 2010) :

i. Microbial spoilage: degradation of food components due to the activity and/or presence of microorganisms
ii. Enzymatic spoilage: undesirable changes due to the activity of enzyme in enzyme catalyzed reactions
iii. Chemical spoilage: irreversible changes such as discoloration due to non-enzymatic chemical reactions between intrinsic food components or the action of environment to the food components (e.g. Maillard browning and lipid oxidation)
iv. Physical spoilage: undesirable changes in the physical structure of the food (e.g. sugar crystallization in preserves, separation of emulsions and collapse of gels).

Early studies acknowledged that microorganism activity is a crucial cause of food spoilage because it can affect to both quality and safety. During propagation process of food spoilage, some physical properties may be observed such as changes in color, taste and smell, etc

(Wilkes et al., 2000). Analytical techniques commonly used to determine quality of food are gas chromatography-mass spectrometry (GC-MS) technique, bacterial and sensory analysis (Zhang et al, 2010). Up to the present, many researchers have tried to push E-nose forward as one of the standard techniques in food industry to evaluate food quality (Berna, 2010). Because the results of an E-nose are normally reported in terms of aroma pattern yielded from the response of chemical sensor array, therefore several scientists tried to correlate E-nose data with the standard techniques. (Blixt & Borch, 1999; O'Sullivan et al., 2003). The E-nose has been applied in a wide range of animal products, including: animal flesh product (e.g. meat (Blixt & Borch, 1999; Panigrahi et al., 2006; Vernat-Rossi et al., 1996; Winquist et al., 1993;), fish and seafood (Alimelli et al., 2007; Huang et al., 2011; Kent et al., 2004; Lorwongtragool et al., 2011; **Ólafsdóttir**, et al., 1997, 2004; Rajamäki et al., 2006; Zhang et al., 2009), dairy product (e.g. cheese and milk) (Ampuero & Bosset, 2003; Drake et al., 2003; Seregély et al., 2006) and egg product (Suman et al., 2007; Yongwei et al., 2009)). Most applications are associated with using E-nose to determine the degree of microbial spoilage.
In the literature, development of E-nose system for food monitoring has been conducted in the same direction. E-nose is expected to be usable for on-line analysis in quality control (Blixt & Borch, 1999; Vernat-Rossi et al., 1996). In 1996, E-nose with a limited number of semiconductor gas sensors (six sensors: TGS822, TGS812, TGS824, TGS825, TGS880 and TGS800) was demonstrated to discriminate cured meat products such as dry sausages of various origins or cured hams of different qualities. It was also used for analysis of the volatile compounds of bacterial strains (Vernat-Rossi et al., 1996). The experiment was done by pumping (10 ml min^{-1}) headspace generated by bacteria after 48 h to a gas sensor array. Fig. 1 shows analysis diagram of the volatile compounds of bacterial strains.

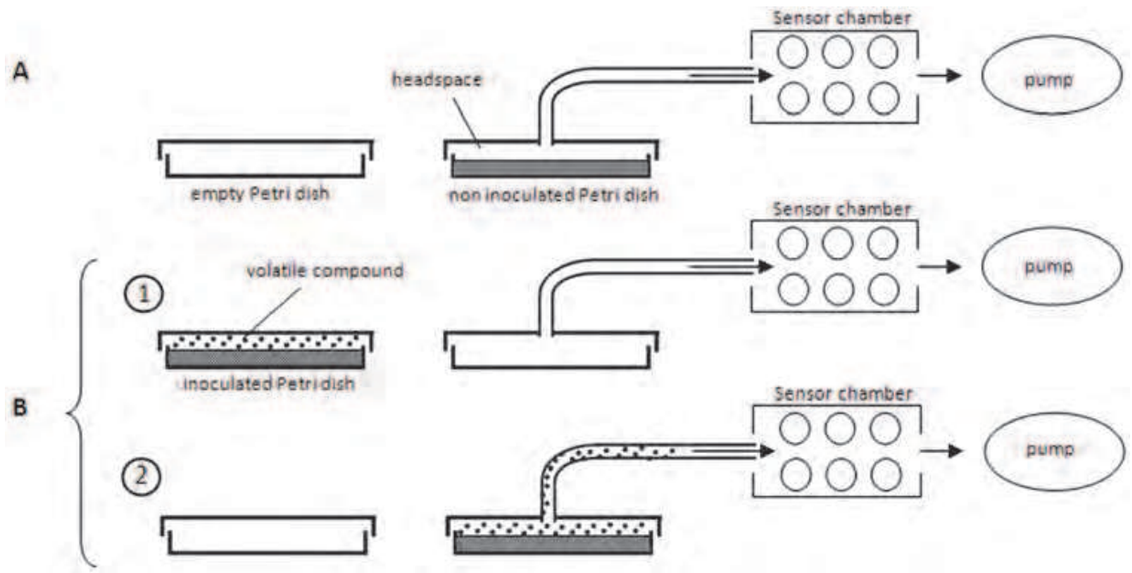

Fig. 1. Analysis of the volatile compounds of bacterial strains. (A) Preliminary analysis to train the gas sensor. Volatile compounds of a non-inoculated Petri dish are introduced for 40 s through the measurement chamber. (B) The two stages of the volatile-colapound analysis: (1) return to baseline for the gas-sensor signal; extraction of the headspace of an empty Petri dish for 10 min; (2) extraction of volatile compounds from an inoculated Petri dish and analysis for 40 s. (Vernat-Rossi et al., 1996)

The performance of E-nose has been demonstrated for quality estimation of ground beef and pork (Winquist et al., 1993). The studies were performed by using a sensory array composed of 10 metal oxide semiconductor field-effect transistors, four Tagushi type sensors and one CO_2 -sensitive sensor. With the same sensor array, Blixt and Borch (Blixt & Borch, 1999) showed that the degree of spoilage of vacuum-packed beef could be determined quantitatively. They also developed mathematical model, describing the relationships between the degree of spoilage, as determined by a sensory panel, and the sensor signal magnitudes of the electronic nose. This relationship is given in equation (2.1)

$$Y = b_0 + b_1 \times S_1 + b_2 \times S_2 + ... + b_n \times S_n \quad (2.1)$$

where Y is the degree of spoilage, b_x is the unweighted regression coefficient obtained from partial least-squares regression (PLS), and S_x is the standardized sensor signal magnitude of the electronic nose. The factor 1/(weight of meat (g))$^{1/2}$ was used as the standardized sensor signal.

It is noticed that most applications for evaluating food quality prefer metal oxide gas sensors as a sensor array (Berna, 2010). Metal oxide semiconductor gas sensors can respond highly and rapidly to volatile compounds. However, this type of gas sensors work at high temperature, thereby consuming higher electrical power than others (Fleischer& Meixner, 1997; Tomchenko et al., 2003) Gas sensors with lower power consumption are therefore in higher demand nowadays. Many research groups have developed alterative sensing materials that can be sensitive to volatile compounds at lower temperature. Hybrid metal oxide and multi-walled carbon nanotube (SnO_2/MWCNTs, WO_3/MWNTs films) was found to present excellent sensitivity towards NO2 at room temperature (Espinosa et al., 2007; Su & Pan, 2011). Conducting polymer as well as polymer/conductive filler composite are new sensing materials that can be applied to indicate quality of food products. Conductive filler such as carbon black, carbon nanotube and graphite can be loaded into the polymer matrix at the level near the conduction percolation threshold to obtain high gain sensors. For example, four nanocomposite sensor materials based on conductive polymer loaded with carbon black were demonstrated for potential applications in real time analysis and quantification of the odor given off from a selection of food borne pathogens including *Salmonella spp.*, *Bacillus cereus* and *Vibrio parahaemolyticus.* (Arshak et al., 2007)

Applications of E-nose involving fish and seafood freshness monitoring were also widely interested. Freshness monitoring of such protein-based products is usually based on chemical contents decomposed by microbial and chemical spoiling process, generally known as total volatile basic nitrogen (TVB-N) such as trimethylamine (TMA), ammonia (NH3) and dimethylamine (DMA). (Dapkevicius et al., 2000; Gram & Huss, 1996; Önal, 2006; Pacquit et al., 2006; Seo et al., 2011;) Because many fish species contain high content of free amino acid and trimethylamine oxide (TMAO), TMA is formed from the reduction of TMAO by bacterial activity (Howgate, 2010a, 2010b).

In order to apply the E-nose efficiently for a specific application, the gas sensor array containing in the E-system should be highly sensitive and selective to the volatile compounds presented in the samples. Lorwongtragool and co-workers have designed a new type of polymer/SWNT-COOH gas sensors that are highly sensitive to the presence of amine based volatile compounds (Lorwongtragool et al., 2011). Fig. 2 shows the sensor responses of polymer/SWNT-COOH nanocomposites to dimethylamine, dipropylamine,

pyridine, and ammonium hydroxide at the concentrations of 50-1000 ppm. This E-nose was shown to be an efficient device for fish freshness monitoring.

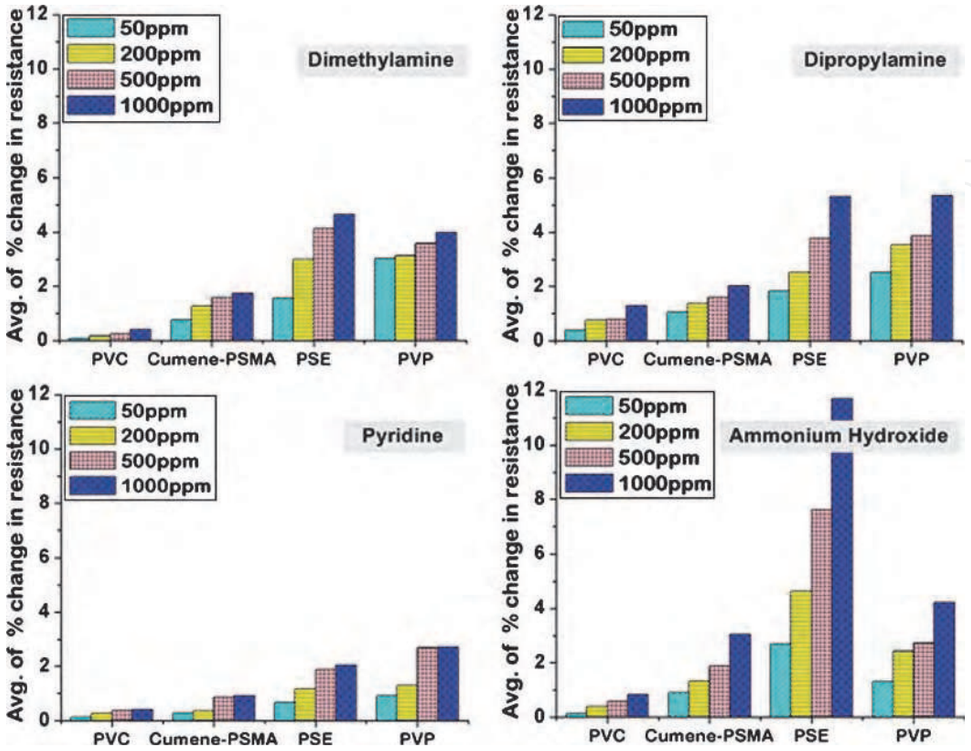

Fig. 2. The average of percent change in resistance of each sensor in static condition when exposed to (a) dimethylamine (b) dipropylamine, (c) pyridine, and (d) ammonium hydroxide at the concentrations of 50-1000 ppm (Lorwongtragool et al., 2011)

2.2 Animal waste

This section illustrates the literature and our recent works on malodor measurement in farms. Problems of most livestock industries often concern with unpleasant odor generated by animal waste resulting in conflict between producers and the local public. In fact, many factors that cause air pollution are associated with farm management (Hamelin et al., 2010;Ivanova-Peneva et al., 2008; Melse & Timmerman, 2009; Sheridan et al., 2002; Van der Werf et al., 2005; Pan et al., 2006, 2007). Numerous animal farms has employed E-nose for odor monitoring such as pig (Brose et al., 2001; Lorwongtragool et al., 2010), poultry (Sohn et al., 2008), sheep (Cramp et al., 2009) and cattle farms (Lane & Wathes, 1998). With complexity of odor in the livestock farm, odor monitoring, reducing and controlling are required due to the impacts of health risks of human and quality of livestock production (Pan & Yang, 2007).

Over 400 compounds identified by GC-MS technique are normally generated from anaerobic process in pig farm area. The compounds identified include many acids, alcohols, aldehydes, amides, amines, aromatics, esters, ethers, fixed gases, halogenated hydrocarbons, hydrocarbons, ketones, nitriles, other nitrogen-containing compounds, phenols, sulfur-containing compounds, steroids, and other compounds (Schiffman et al., 2001). Although analytical instruments such as gas chromatography and mass spectrometry can identify

individual components in a complex odor, the data obtained are lacking the qualitative characteristics as perceived by the human nose (Hyung et al., 1997). Moreover, GC-MS technique as well as other traditional technique (e.g. human assessment) does not support the monitoring on site and at real time due to the method of sampling. Schiffman, et al. (Schiffman et al., 2001) collected the odor onto Tenax® cartridges and Tedlar® *bags* to be analyzed by GC-MS. Odor threshold and sensory assessments were carried out by human assessors. The flexibility of E-nose has shown its ability beyond the human panels with its long operating hours and good reproducibility. Since E-nose containing air flow unit in the system can suck the air directly into the sensing unit, it can be a better choice than taking air into a gas sample bag that posses many difficulties (Trabue et al., 2006). Fig. 3 shows schematic diagram of the portable E-nose system using for detection of unpleasant odor in pig farm (Lorwongtragool et al., 2010).

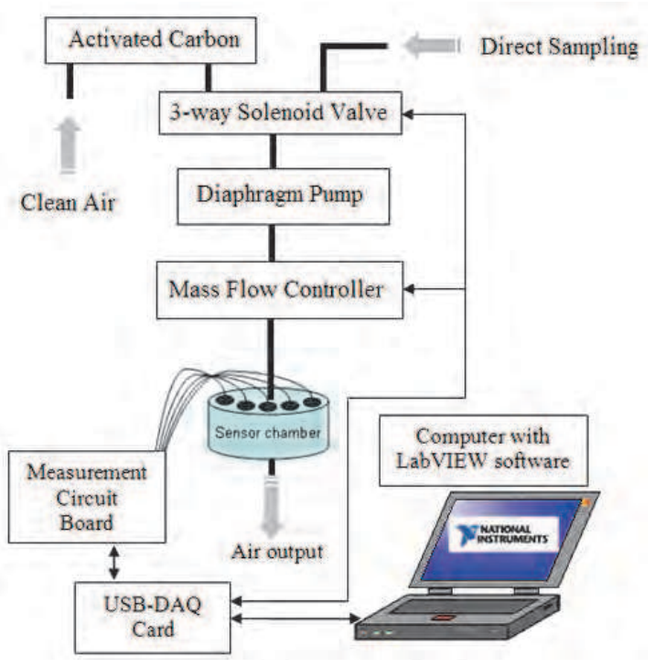

Fig. 3. Schematic diagram of the portable e-nose system (Lorwongtragool et al., 2010).

The odor measurement by using E-nose works under switching between the reference gas (purified air) and odorous gases (non-purified air) pumped into the sensor chamber from the point of sampling. The reference gas was obtained from purification of air with activated carbon. Direction and rate of flow are controlled by 3-way solenoid valve and a mass flow controller, respectively. The USB-DAQ card used as interface device sends analog signal for flow rate adjustment and receives a set of digital signal back for reading the real-time flow rate value. Odor can be directly sampled from interested point by pumping. Because it is well-known that the air inside a pig farm concerning usually consists of ammonia and hydrogen sulfide (Kim et al., 2008; Timmer et al., 2005), so the gas sensor array should be chosen appropriately to detect these gases.

Polyethylene odor sampling duct may be constructed to provide well-mixed (homogenous) air samples. Sohn et al. (Sohn et al., 2008) used a temporary duct with a design of Australian Standard AS 4323.1 as shown in Fig. 4. The complex odour-generating mechanisms within

poultry housing were continuously monitored by E-nose based on 12 metal oxide semiconductor sensors.

Fig. 4. Tunnel ventilated broiler shed showing polyethylene sampling duct attached to an exhaust fan. (Sohn et al., 2008)

Actually, the E-nose used to monitor the odor in the farm area is useful for supporting farm management. Numerous published papers have used the E-nose to study the parameters relating to changes of malodorous odors and used as one of the equipment to find out the final solution for odor reduction. According to Lorwongtragool et al. (Lorwongtragool et al., 2010), the E-nose based on eight commercial metal oxide gas sensors used as assessment technique finally led to an optimized feed menu and cleaning schedule to control and reduce the odor in pig farm. Environmental conditions such as season, temperature, wind, and humidity, etc. that are difficult to control and manage bring the emitted odor to spread to neighbor areas have been evaluated by E-nose (Pan et al., 2006, 2007). In addition, E-nose with applying analysis software can highly improve the accuracy and precision of livestock farm odor evaluation.

Recently, the non-specific conducting polymer was used as chemical gas sensing materials in E-nose system for monitoring odor emissions from a biofiltration system in a piggery building (Sohn et al., 2009). Based on PCA analysis the odor samples collected at the outlet of the biofilter and those from the inlet and post-humidifier were discriminated. Data pre-processing techniques including normalising and outlier handling could also enhance the odor discrimination performance.

E-nose can also be used to detect illness of animals (Cramp et al., 2009; Lane & Wathes, 1998). There are only a few published papers reported about using E-nose to diagnose or detect unusualness of animal; for examples, detection of perineal odors associated with oestrus in the cow (Lane & Wathes, 1998) and detection of cutaneous myiasis in sheep (Cramp et al., 2009). In early stage, the flystrike (Cutaneous myiasis) is difficult to detect,

therefore cutaneous myiasis causes the death of millions of sheep each year due to the larvae of flies burrowing into and feeding on body tissues (Krajewski et al., 2009; Lane & Wathes, 1998; McGraw & Turiansky, 2008). Since a characteristic unpleasant odor is generated from flystrike, this leads to a potential application of E-nose technology to detect the flystrike in advance.

The E-nose combined with non-linear signal measurement techniques and linear discriminant analysis (LDA) extracts signal features and process those features for classification of odor groups. Schematic diagram of the experimental system is given in Fig. 5 (Lane & Wathes, 1998). The E-nose system composed of six metal oxide gas sensors chosen on the basis of GC-MS profile of flystrike odor. Each of the odor sample and clean air is drawn through the E-nose chamber by using diaphragm vacuum pump. The direction of air flow is controlled by two solenoid valves to switch between sample odor and clean air. This experiment can accurately distinguish flystrike odor on days 1, 2 and 3 of development from that of dry wool in all experiments ($P < 0.05$).

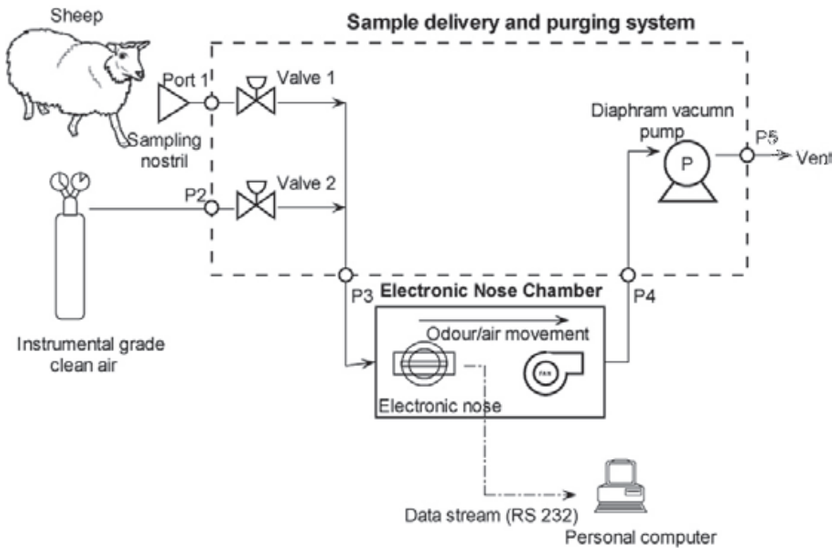

Fig. 5. Schematic diagram of the experimental system, which included the odor or clean air source, the sensor array enclosed in an airtight chamber, an odor delivery system, moisture trap and computer for data logging and recording (Cramp et al., 2009)

3. Human body odor

3.1 Historicatl background

Human body odor refers to unpleasant and pleasant scents that can be emitted from many parts of the body such as skin, armpit, mouth, feet, etc. In fact, most of body odors were considered socially unpleasant while there are only a few of them that are believed to serve as attractants. The human body odor was first investigated systematically in 1936 (Yaglou et al., 1936). They studied the perceived air quality on different numbers of persons who were seated in an experiment-controlled room. They found the relationship of ventilation rates per person required to provide a certain acceptable odor intensity as emitted from the human body in a definite space. Their work has then become the basis of European guidelines for ventilation rates in buildings for more than 50 years.

However, during that time, no scientific instrument existed to measure perceived air quality or human body odor directly. Thus, human panel test, assisted by statistical analysis, had been the only mean to evaluate perceived air quality. The perceived air quality (AQ) in decipols can be written down as (Fanger, 2001):

$$AQ = 112\left[\ln\left(\frac{\exp(-0.18-5.28\overline{AA})}{1+\exp(-0.18-5.28\overline{AA})}\times 100\right)-5.98\right]^{-4} \tag{3.1}$$

Where $\overline{AA}$ is the mean vote of air acceptability (from -1 to +1) measured from untrained panel of impartial subjects who enter the space and judge the acceptability of the air quality.

Nowadays, analytical instruments such as gas chromatograph–mass spectrometry (GC-MS) and electronic nose (E-nose) have become available for detection and analysis of human body odor with both qualitative and quantitative capabilities. Human body odor consists of various volatile organic compounds (VOC). By using stir bar sorptive extraction in connection with thermal desorption, Penn and co-workers (Penn et al., 2007) found on average 241 peaks in the GC–MS of armpit sweat, 179 in saliva and 163 in urine per individual. The list of 152 identified VOCs in the GC–MS profiles from armpit sweat is displayed in Table 1.

RT (min)	**Identification**
	alcohols and phenols
9.98	2-phenylethanol
12.3	a-terpineol
12.47	γ-terpineol
12.9	2-phenoxyethanol
13.34	citronellol
13.82	geraniol
16.48	eugenol
18.93	isoeugenol
21.93	1-tridecanol
26.83	pentadecanol
27.75	a hexadecadienol
29.51	hexadecanol
	Aldehydes
12.74	decanal
13.93	p-anisaldehyde
14.38	geranial
15.46	undecanal
18.1	dodecanal
20.61	tridecanal
20.83	lilial
23	tetradecanal
23.58	pentylcinnamaldehyde
26.04	E-2-hexylcinnamaldehyde
28.09	hexadecanal

RT (min)	Identification
	Ketones
8.98	acetophenone
9.49	2-nonanone
14.99	2-undecanone
15.42	an isopropylacetophenone
17.47	jasmone
17.69	2-dodecanone
19.45	γ-irone
19.66	β-ionone
20.22	2-tridecanone
20.54	Z-α-irone
22.52	2-tetradecanone
18.27	a-ionone
23.09	benzophenone
24.08	1-ethyl-3-methyl-β-ionone
24.92	2-pentadecanone
27.54	2-hexadecanone
28.7	2-acetyl-3,5,5,6,8,8-hexa-methyl-5,6,7,8- tetrahydronaphthalene
29.22	7-acetyl-6-ethyl-1,1,4,4-tetramethyl tetralin (Musk 36A)
	Esters
11.4	benzyl acetate
12.07	2-phenylethyl acetate
12.73	dihydromyrcenol acetate
14.82	cis-2-tert-butylcyclohexyl acetate
16.37	a-terpinyl acetate
16.43	citronellol acetate
16.62	neryl acetate
17.16	geranyl acetate
17.86	methyl-N-methylanthranilate
18.28	2-hexyl-2-pentenoate
18.79	E-cinnamyl acetate
21.24	a-trichloromethylbenzyl acetate
21.95	pentyl salicylate
22.21	isooctanedioldibutyrate
22.69	isoeugenol acetate
23.3	isopropyl dodecanoate
23.73	methyl-cis-dihydrojasmonate
24.15	3Z-1-hexenyl salicylate
24.46	1-hexyl salicylate
25.01	methyl trans-jasmonate
25.73	a hexenyl salicylate
26.65	benzyl benzoate
27.38	ethyl tetradecanoate
27.53	2-ethylhexyl salicylate
28.23	a branched isopropyl hexadecanoate

RT (min)	Identification
30.06	ethyl pentadecanoate
30.32	2-phenylethyl phenylacetate
30.93	methyl palmitate
32.29	decyl octanoate
32.36	dodecyl hexanoate
32.4	hexyl dodecanoate
32.99	ethyl hexadecanoate
33.7	isopropyl hexadecanoate
35.64	ethyl heptadecanoate
36.47	a branched dodecyl benzoate
37.35	2-ethyl-hexyl 4-methoxycinnamate
38	dodecyl octanoate
38.72	dodecyl benzoate
43.19	tridecyl benzoate
47.56	tetradecyl octanoate
49.15	tetradecyl benzoate
	Hydrocarbons
7.67	p-cymene
12.26	1-dodecene
12.5	dodecane
15.15	tridecane
17.55	1-tetradecene
17.77	tetradecane
18.22	β-caryophyllene
19.72	trans-muurola-4(14),5-diene
19.96	a methyl biphenyl
20.36	α-farnesene
20.38	pentadecane
21.77	4-methylpentadecane
22.56	hexadecane
23.28	5-phenylundecane
23.53	4-phenylundecane
23.99	3-phenylundecane
24.94	2-phenylundecane
25.04	heptadecane
25.49	6-phenyldodecane
25.66	a sesquiterpene
25.77	5-phenyldodecane
26.96	a propyl-substituted dodecane
27.48	octadecane
27.57	2-phenyldodecane
28.09	6-phenyltridecane
28.64	4-phenyltridecane
29.4	3-phenyltridecane
29.54	3-methyloctadecane

RT (min)	Identification
30.13	1-nonadecene
30.22	nonadecane
30.38	2-phenyltridecane
32.19	3-methylnonadecane
32.93	eicosane (C-20 linear hydrocarbon)
35.79	heneicosane (C-21 linear hydrocarbon)
38.79	docosane (C-22 linear hydrocarbon)
41.49	tricosane (C-23 linear hydrocarbon)
43.44	tetracosane (C-24 linear hydrocarbon)
	Amines
12.65	2-pentylpyrrole
13.28	2-phenoxyethylmethylamine
15.19	an aliphatic amine
16.34	nicotine
16.81	4-sec-butylaniline
20.37	N,N-dimethyl-1-dodecylamine
30.24	N,N-dimethyl-1-hexadecylamine
35.88	N,N-dimethyl-1-octadecylamine
	Amides
17.18	methyl N,N-diethylthiocarbamate
18.04	a hydroxy acetanilide
21.98	n-propylbenzamide
	Carboxylic acids
12.24	octanoic acid
14.76	nonanoic acid
21.03	8-methylundecanoic acid
21.9	dodecanoic acid
22.8	a methyl dodecanoic acid
23.18	9-methyldodecanoic acid
25.74	10-methyltridecanoic acid
26.8	myristic acid (tetradecanoic acid)
28.36	a methyltetradecanoic acid
28.65	a methyltetradecanoic acid
28.84	9-pentadecenoic acid
29.31	pentadecanoic acid
30.76	a methylpentadecanoic acid
31.7	9-hexadecenoic acid
32.49-33.00	palmitic acid (hexadecanoic acid)
34.37	9-heptadecenoic acid
37.31	oleic acid
38.03	stearic acid (octadecanoic acid)
	Lactones
12.27	γ-heptanolactone
16.7	γ-nonanolactone
18.6	coumarin

Table 1. Compounds identified from armpit samples (Penn et al., 2007).

The changes in body odor associated with aging were investigated by Haze and co-workers (Haze et al., 2001). They found that 2-Nonenal compound only presents in the odor of individuals over the age of 40 years. The 2-Nonenal compound and other aldehydes were produced from oxidative degradation of monosaturated fatty acids such as palmitoleic acid and vaccenic acid. Some VOCs associated with aging that were detected by GC/MS are displayed in Table 2.

Compounds	**Detection rate (%)**	
	<40 y (n=9)	>40 y (n=13)
Hydrocarbons		
1-Octene	11	8
Decane	11	23
Undecane	22	23
Dodecane	67	69
Alcohols		
1-Butanol	11	8
1-Hexanol	11	15
2-Ethylhexanol	89	85
Octanol	11	8
1-Decanol	11	15
Amyl alcohol	11	8
Hexadecanol	11	8
Octadecanol	11	8
Acids		
Acetic acid	22	23
Butyric acid	22	15
Ketones		
4-Methyl-2-pentanone	11	8
6-Methyl-5-heptenone	89	77
Aldehydes		
Hexanal	33	23
Heptanal	11	15
Octanal	89	85
Nonanal	89	85
Decanal	89	69
2-Nonenal	0	69

Table 2. Some compounds detected from body odor by GC/MS (Haze et al., 2001).

Comparison of compounds extracted from the samplings of male and female was also studied by Curran and co-workers (Curran et al. 2005a). Some compounds such as dodecanoic acid, propanedioic acid-methyl ester, octanal, and tetradecanoic acid could be extracted from the male subjects only and they were not present in any of the female profiles. Overall, the individual human body odors can be determined by several factors, which are either stable over time (genetic factors) or vary with environmental or internal conditions. The human body odor can thus be classified into three types (Curran et al. 2005b).):

i. "Primary odor" of a person contains constituents that are stable over time regardless of diet or environmental factors.

ii. "Secondary odor" contains constituents that are present due to diet and environmental factors.
iii. "Tertiary odor" contains constituents that are present because of the influence of outside sources (i.e., lotions, soaps, perfumes).

For identification of an individual using the human body odor, detection of the primary odor class should be used because they are stable over time and diverse across people.

3.2 Evaluation of body odor strength

There are more refined and less subjective ways to measure odor strength in direct way. For instance, the concept of dilution-to-threshold principle can be used quite accurately to reduce uncertainties associated with subjective impressions (Nicell, 2003; Henshaw et al., 2006; Kim & Park, 2008). In the cosmetic industry, human olfaction has been commonly employed to evaluate the odor strength of armpit for the development of deodorants. The armpit odor comprises a complex set of chemicals as shown in section 3.1. However, to simplify the odor strength of armpit, only a single component such as isovaleric acid can be used for training the sensory panel (Hooper et al., 1982). Isovaleric acid (3-Methylbutanoic acid) is a natural fatty acid that can represent the sweaty primary odor (Amoore, 1967, Leyden et al., 1981) which contributes mainly to the armpit malodor. Molecular structure of isovaleric acid is shown in Fig. 6.

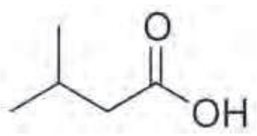

Fig. 6. Isovaleric acid ($(CH_3)_2CHCH_2COOH$)

Hooper and co-workers (Hooper et al., 1981, 1982) assigned the concentrations of isovaleric acid levels on a scale 0 to 5 corresponding to subjective impression by human nose, as shown in Table 3.3. Their test was carried out by a team of three female assessors of ages ranging from 20 to 40 years. They were selected for olfactory evaluation on the basis that each person is able to rank correctly the odor levels of the series of aqueous isovaleric acid solution listed in Table 3.

Level	Concentration of aqueous isovaleric acid solution (mM)	Subjective impression
0	0	No odor
1	0.12	Slight
2	0.48	Definite
3	1.99	Moderate
4	7.88	Strong
5	32.33	Very strong

Table 3. The concentration of the isovaleric acid levels that correspond to subjective impression using human nose.

Recently, evaluation of body odor strength based on isovaleric acid solutions as prepared according to the intensity scale can also be performed using gas sensor array in stead of the

human nose (Wongchoosuk et al., 2009a). The response of a gas sensor array to the isovaleric acid is displayed in Fig. 7.

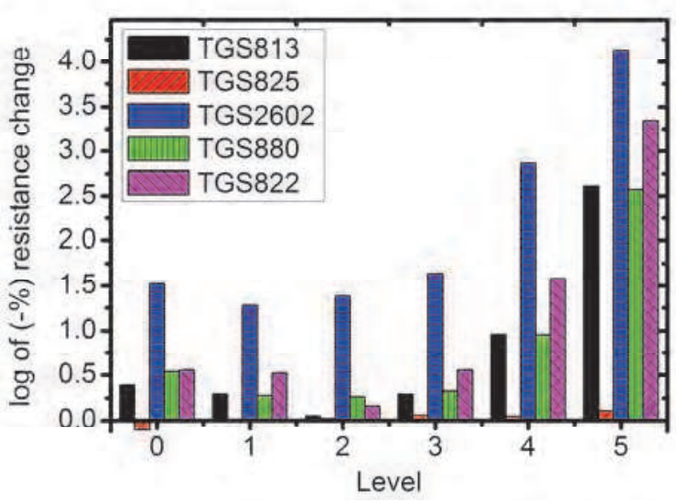

Fig. 7. Logarithmic plot of the sensor response to isovaleric acid at different intensity level (Wongchoosuk et al., 2009a).

3.3 Instrument and method for real-time human odor detection

Even though GC-MS method can be used to detect VOCs emitted from human body odor, they are quite time-consuming, complicated and so costly. Therefore, it is impractical to use it in real-time or at point of use, especially where quick screening for detecting human body odor is the case. One of the most state-of-the-art technologies that can be used for real-time human odor detection is electronic nose (E-nose). Recently, there have been increasing interests in the application of E-nose for measurement of human body odors. If successful, many new applications await in such area as healthcare monitoring, biometrics, homeland security and cosmetics. The first success in distinguishing between two different people by detecting human odor from the armpit region using E-nose was reported by Wongchoosuk and co-workers (Wongchoosuk et al., 2009a). Schematic diagram of the E-nose system for real-time human odor detection is exhibited in Fig. 8.

To allow an identification of human odors, principal component analysis (PCA) was employed to perform pattern recognition and discrimination. PCA is a popular statistical technique usually used to visualize in two or three uncorrelated dimensions transformed from all correlated information. In principles, PCA process contains five following steps (Wongchoosuk et al., 2009a):

i. Get data from matrix, $X_{M\times N}$. The row M represents different repetition of the experiment and the column N represents the number of independent sensors.
ii. Normalize the data matrix, $Norm(X_{M\times N})$, by the mean subtraction. The mean of each N column is calculated and subtracted from the data set. Hence, the new data set produces the mean equal to zero.
iii. Calculate the covariance matrix, $Cov(X_{M\times N})$, and calculate eigenvectors and eigenvalues of the covariance matrix. The calculated eigenvectors must be unit eigenvectors.

iv. Rearrange the eigenvectors and eigenvalues. The eigenvectors are ordered by eigenvalues from highest to lowest, $(\overrightarrow{Cov(\mathrm{X}_{M\times N})})_{\max\to\min}$.

v. Obtain the PCA result by matrix multiplication and transpose, $\left((\overrightarrow{Cov(\mathrm{X}_{M\times N})})_{\max\to\min} \otimes Norm(X_{M\times N})\right)^{T}$. The obtained new dataset with orthogonal linear transformation have been plotted in two or three dimensions containing the most relevant of the data set.

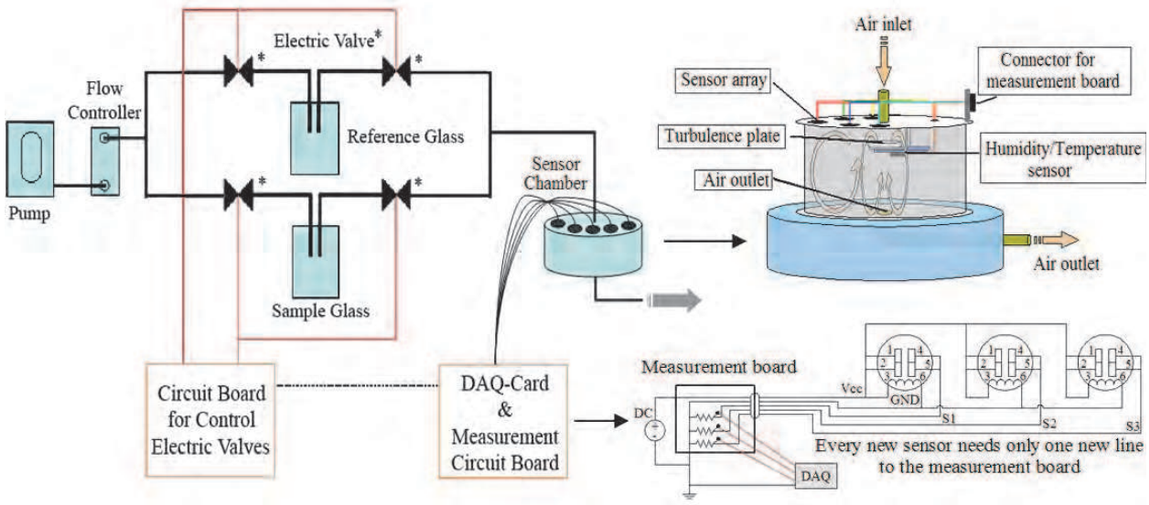

Fig. 8. Schematic diagram of the E-nose system (Wongchoosuk et al., 2009a).

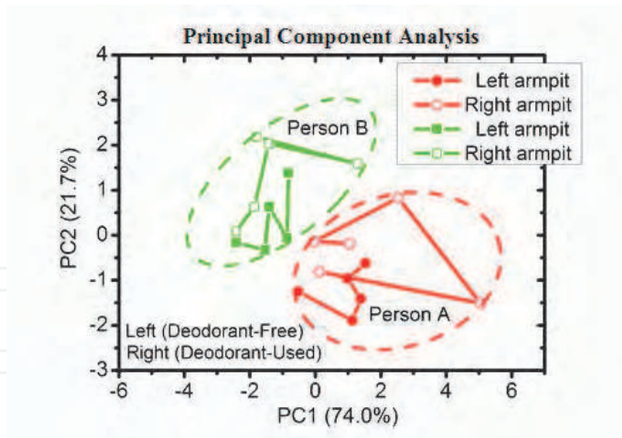

Fig. 9. Discrimination of two persons (Wongchoosuk et al., 2009a).

By using typical E-nose with PCA, Wongchoosuk and co-workers showed obviously discrimination between person A and person B and the use of deodorant may not change the odor fingerprint (Fig. 9). To recognize and discriminate more persons, the method for sensor signal correction should be developed. It is well-known that sensor drift effect is a serious impairment of chemical sensors. The sensor signals alter over time. Therefore, they will produce different responses for the same odor. To correct the sensor drift effect, a mathematical model has been applied to the raw sensor signal via the following formulation:

$$S'(t) = S(t) - F(t) + \overline{F(t)} \tag{3.2}$$

Where $S'(t)$ represents the corrected sensor signal. $S(t)$ is the smoothed raw sensor signal. $F(t)$ is defined as spline interpolation calculated from relationship between the averaged values of their 10 neighboring data points that switch from reference to sample of each loop. $\overline{F(t)}$ is a mean valve of $F(t)$. The Eq. 2 can be demonstrated in Fig. 10.

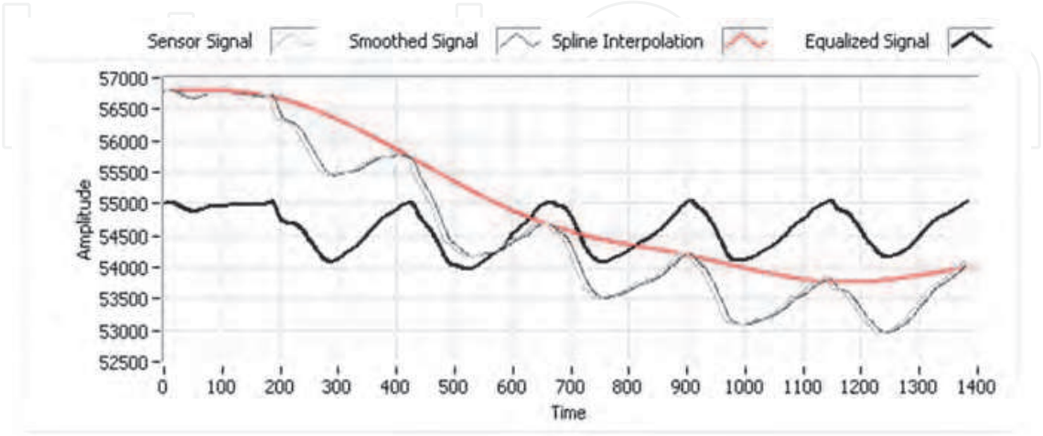

Fig. 10. Correction of sensor drift.

The real armpit measurement by E-nose is shown in Fig. 11. It clearly shows the sensor drift effect that makes the difference of the baseline shift, especially the signal from sensor 1 and

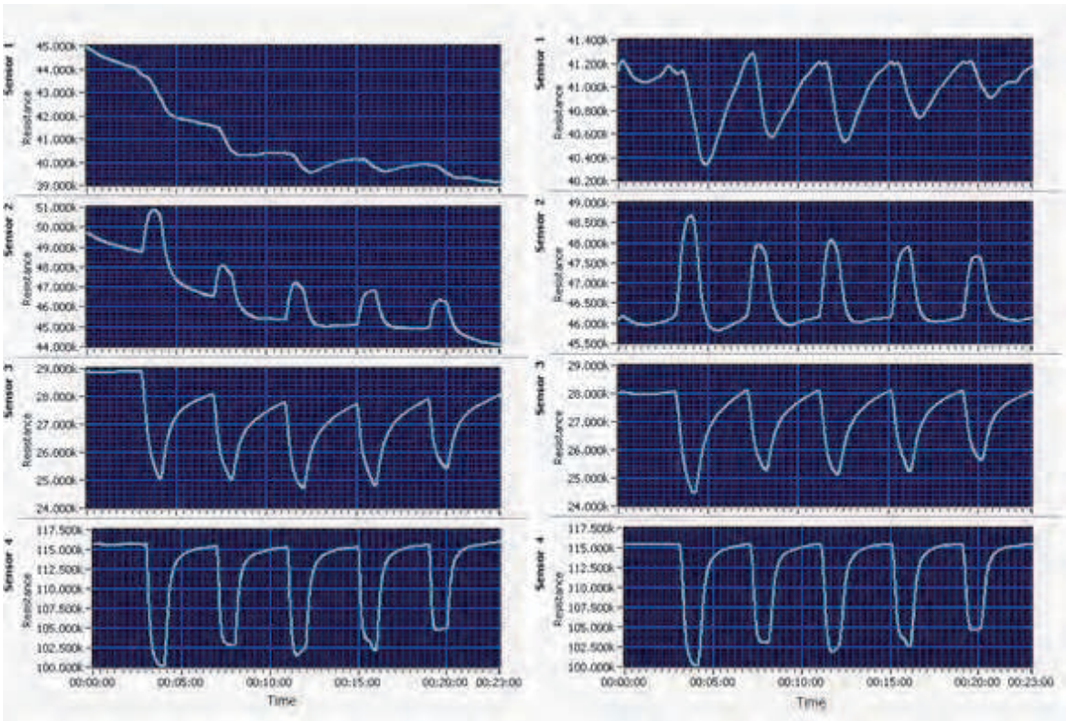

Fig. 11. Raw sensor signals (L-hand side) and corrected sensor signals by using mathematical model (R-hand side).

sensor 2. The drift effect of sensor signal may arise from the temperature variation under long time measurement and humidity generated from armpits (sweat) during the measurement. After correction by using the mathematical model, the sensor signal becomes more homogeneity. It will be useful for reorganization and discrimination of many persons because it helps to reduce the humidity and temperature effects.

To add 95% confidence ellipse for an XY scatter plot in PCA results, the equation of an ellipse is given by the following equation:

$$\frac{(X-X_0)^2}{a^2}+\frac{(Y-Y_0)^2}{b^2}=1 \tag{3.3}$$

$$\text{Where } a=\sqrt{\frac{\sum_i^N x_i^2}{N}} \text{ and } b=\sqrt{\frac{\sum_i^N y_i^2}{N}} \tag{3.4}$$

The X_0 and Y_0 can be calculated from the center of mass of XY scatter plot. The ellipse is rotated from the horizontal by the following angle:

$$\theta=0.5\tan^{-1}\left(\frac{2\sum_i^N(x_i-\bar{x})(y_i-\bar{y})}{\sum_i^N(y_i-\bar{y})^2-\sum_i^N(x_i-\bar{x})^2}\right) \tag{3.5}$$

The PCA result shows clear classification of four persons within 95% confidence ellipses as shown in Fig .12. The results confirm that each human body has a unique odor pattern. Even though odor of each person can be changed under diverse living conditions such as eating, drinking, sexual activities, health or hormonal status, the E-Nose is still able to identify the people from armpit odor region.

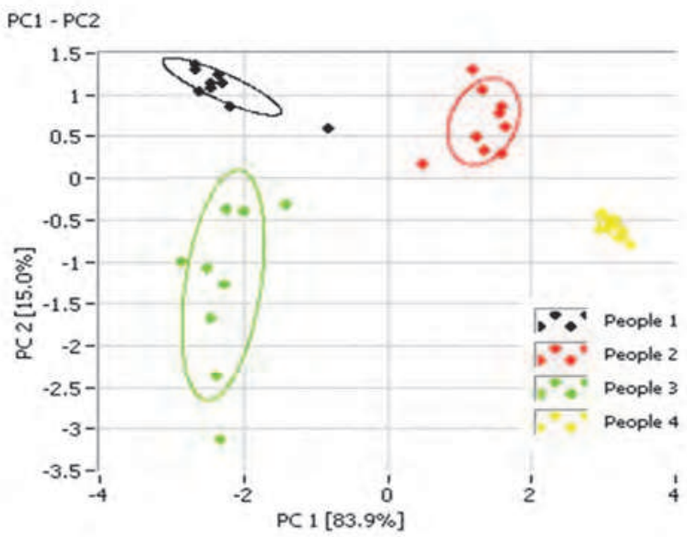

Fig. 12. Discrimination of four persons.

Based on the results as shown above, it is expected that E-nose has potential for helping identify terrorists from a distance in the near future.

4. Indoor air monitoring

Most people spend more than 80% (90% in industrial countries) of their time indoors (Austin et al., 1992), i.e., in offices, houses, stores, restaurants, public or private transportation vehicles, movie theatre, etc. Typically, several hundreds of indoor chemical contaminants including by-products of the combustion (CO_2 , CO), cigarette smoke, particulate matter, mineral fibers etc. can be found. A list of typical indoor air pollutants is displayed in Table 4. In spite of the very low concentrations, some of these compounds are extremely toxic such as CO, NO_2 or formaldehyde. Only as low as 667 ppm of CO may cause up to 50% of the body's hemoglobin to convert to carboxyhemoglobin (Tikuisis et al.,1992) that is ineffective for delivering oxygen to bodily tissues. Exposure to 100 ppm of NO_2 can produce pulmonary edema that may be fatal or may lead to bronchiolitis obliterans while formaldehyde was proved to be carcinogenic. Concisely, indoor air quality can greatly affect morale, emotion, productivity, and health status of people. Therefore, development of technical devices for indoor air monitoring has become an important issue of public interest. In 1988, Fanger (Fanger, 1988) proposed a method for assessing the air quality and introduced that discomfort as caused by indoor air quality based on human sensory panels. However, his method is too time consuming and cannot be used for continuous measurements in long time monitoring and control.

Inorganic Pollutants	Organic Pollutants	Physical Pollutants	Biological Pollutants
Carbon dioxide	Volatile Organic Compounds	Particulate matter	House dust mites
Carbon monoxide	Formaldehyde	Asbestos	Dander from furred animals
Nitrogen dioxide	Pesticides	Radon	Fungi
Sulphur dioxide	Polynuclear Aromatic hydrocarbons		Bacteria
Ozone	Polychlorinated biphenyls		

Table 4. Typical indoor air quality contaminants

In the last decades, gas sensor systems (E-nose) have been developed for monitoring air quality instead of human sensory panels. In an E-nose system, the gas sensors may be considered the most important component. For developing individual gas sensor, there is a great deal of effort on improving the sensitivity and selectivity using nanotechnology. A list of nanostructure gas sensors for indoor chemical contaminant monitoring is given in Table 5.

In many cases, individual gas sensor, that provides only one output signal, is not sufficient for monitoring a wide range of gases. The combination of several gas sensors yielding an E-nose is therefore necessary to measure, maintain and control indoor air quality in real-world applications. Unfortunately, typical commercial E-nose architectures are not suitable for such usage. Most of them are usually designed for fixed location that relies on wired connectivity. Moreover, they are power consuming and very limited in local data processing

capabilities (Vito et al., 2008). Therefore, design of new E-nose architectures such as networked or wireless E-nose is a current interesting topic of E-nose development for indoor air monitoring.

Pollutants	Detection range	Sensing material	Ref.
CO	5–1200 ppm	CuO Nanowires	(Liao et al., 2009)
CO_2	500-2500 ppm	La-SnO_2	(Marsal et al., 2003)
NO_2	5-500 ppb	In_2O_3 Nanowires	(Zhang et al.,2004)
SO_2	2-32 ppm	SnO_2−NiO	(Hidalgo et al.,2005)
O_3	0.2-0.4 ppm	WO_3	(Vallejos et al., 2007)
VOCs	100-1000 ppm	Au-ZnO	(Wongchoosuk et al., 2009b)
CH_2O	0.8-12 ppm	NiO	(Lee et al., 2007)
Pesticide	0.1-1.0 ppm	SnO_2	(Huang et al., 2003)

Table 5. List of nanostructure gas sensor responding to some indoor air quality contaminants.

4.1 Typical E-nose

In principles, an E-nose consists of three main parts: (i) air flow system, (ii) detection system, and (iii) control and data analysis system. The air flow system refers to the way to deliver aroma molecules into the detection system. There are two main types of flow systems, including static and dynamic flow systems. The static system has no vapor flow around the gas sensors and the gas sensors are exposed to vapor at a constant concentration. In the dynamic system, E-nose is subjected to continuous flow of vapor with controllable flow rate during measurement. The simple gas sensor chambers for static and dynamic systems are displayed in Fig. 13.

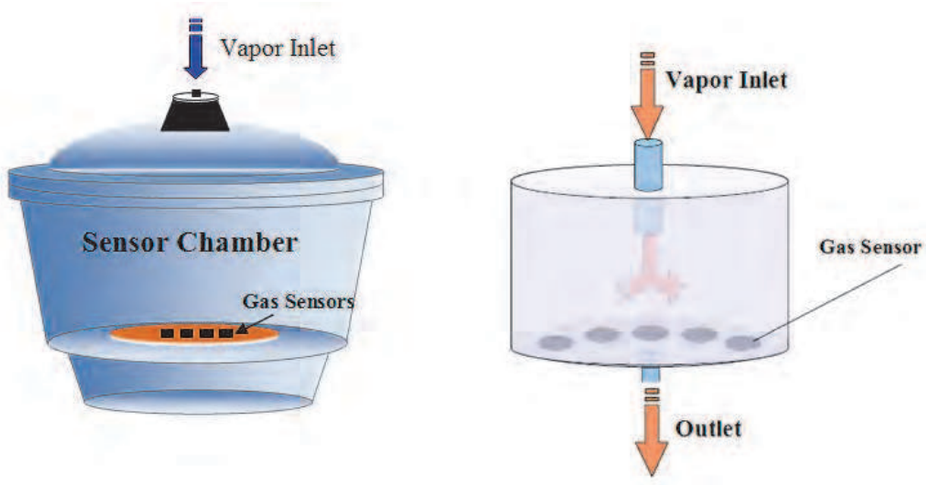

Fig. 13. Static (L-side) and dynamic (R-side) sensor chambers for static and dynamic systems, respectively.

The air flow rate in a dynamic E-nose system is usually controlled by a mass flow controller. Mass flow rate ($\dot{m}$) can be calculated from the density of the gas or liquid (ρ), its velocity (v), and the cross sectional area (A) of the flow by the following equation;

$$\dot{m} = \rho v A \tag{4.1}$$

For an ideal compressible gas, the equation of mass flow rate (Benson, 2008) can be written down as;

$$\dot{m} = \frac{pA}{\sqrt{T}}\sqrt{\frac{\gamma}{R}}M\left(1+\frac{\gamma-1}{2}M^2\right)^{-\frac{\gamma+1}{2(\gamma-1)}} \tag{4.2}$$

Where p is total pressure, T is total temperature, R is gas constant, γ is specific heat ration and M is mach number.

A detection system consists of a gas sensor array embedded in a sensor chamber. There are many types of gas sensors used in E-nose such as metal oxide semiconductors (MOS), conducting polymers (CP), quartz crystal microbalance (QCM), surface acoustic wave (SAW), etc. Each type has specific gas detection principle. The MOS and CP rely on the change in electrical conductivity of the sensing materials for detecting aroma molecules. MOS is usually operated under high temperature. When it is heated, oxygen will be adsorbed onto the sensing surface with a negative charge. Then, donor electrons in the crystal surface are transferred to the adsorbed oxygen, resulting in leaving positive charges in a space charge layer. Thus, the surface potential is formed to serve as a potential barrier against electron flow. Electrical conductivity of MOS is low when it presents in pure air. In the presence of a deoxidizing gas or malodor, the surface density of the negatively charged oxygen decreases, so the barrier height in the grain boundary is reduced. The reduced barrier height results in increasing conductivity of MOS. In a CP gas sensor, swelling of vapor molecules into the polymer film is the basis of sensing mechanism. The swelling decreases the number of connected pathways of the conducting component of the composite material leading to an increase in the electrical resistance of the film. For a pure CP, insertion of analyte molecule into polymer matrix generically increases interchain distance that affects the electron hopping between different polymer chains (Bai & Shi, 2007). The interchain electron transfer can be described by the following relationship (Vercelli et al., 2002):

$$\left(\ln\frac{\sigma}{\sigma_0}\right)^{-1} = \left(\frac{\varepsilon_p}{\left(\varepsilon_s-\varepsilon_p\right)B}\right)X^{-1} + B^{-1} \tag{4.3}$$

Where σ and σ_0 are the conductivity before and after exposure to solvent vapor, respectively. ε_p and ε_s are the relative permittivity of the solvent and the polymer, respectively. B is a constant and X is the molar fraction of absorbed vapor for sensing polymer. A simple linear circuit, called as voltage divider (see Fig. 14), can be used for basic measuring of the resistance of MOS or CP gas sensor array.

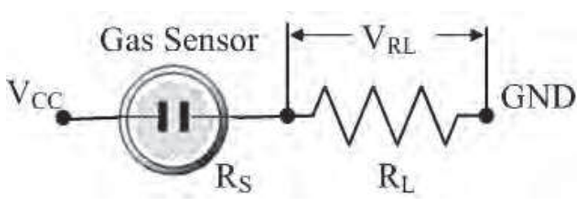

Fig. 14. Basic measuring circuit for MOS or CP gas sensor.

Sensor resistance (R_s) can be calculated by the following equation:

$$R_s = \left(\frac{V_{cc}}{V_{RL}} - 1 \right) \times R_L \tag{4.4}$$

For QCM and SAW gas sensors, a change in the mass of the piezoelectric sensor coating due to gas absorption results in a change in the resonant frequency on exposure to a vapor (Arshak et al., 2004). The QCM utilizes bulk acoustic wave traveling through the entire bulk of the crystal while the SAW uses surface acoustic wave that propagates along the surface of the crystal at a depth of one wavelength at operating frequencies between 100 and 400MHz (Pearce et al., 2003). The resonant frequency of the QCM sensor is related to the change of the mass of QCM loading by the following equation (Sauerbrey, 1959):

$$\Delta f = \frac{-2 f_0^2 \Delta m}{A \sqrt{\rho_q \mu_q}} \tag{4.5}$$

Where Δf is the change in resonant frequency, f_0 is resonant frequency, Δm is mass change due to adsorption of vapor, A is the piezoelectrical active crystal area, ρ_q is density of quartz and μ_q shear modulus of quartz.
In case of the SAW sensor, the change in frequency with sorption of a vapor is given by (Pearce et al., 2003):

$$\Delta f_{SAW} = \frac{\Delta f_p c_v K_p}{\rho_p} \tag{4.6}$$

Where Δf_p is the change in frequency caused by the membrane, c_v is the vapor concentration, K_p is the partition coefficient and ρ_p is the density of polymer membrane.
Both air flow system and detection system are normally controlled by computer via USB, RS-232 or parallel port. Pattern recognition and machine learning such as artificial neural networks (ANN), linear discriminant analysis (LDA), support vector machine (SVM), and principal components analysis (PCA) are typically used in data analysis on a computer.

4.2 Networked E-nose

Networked E-nose can be developed based on a typical E-nose by modifying only one main part (control and data analysis system). If an E-nose can work and analyze the results over a network system such as LAN, WiFi or ZigBee, the E-nose can be defined as networked E-nose. Development of a networked E-nose for indoor air monitoring is demonstrated below:

4.2.1 Air flow system

Schematic diagram of an air flow system for networked E-nose is displayed in Fig. 15.
The measurement works under switching between the reference gas (clean air) and indoor air (malodor) pumped into the network E-nose from the point of sampling. The reference gas was generated by purifying the sucked-in air with activated carbon. The flow was controlled by a mass flow controller and 3-way solenoid valve. Switching between the clean air and the indoor air was used to obtain the baseline and signal, respectively.

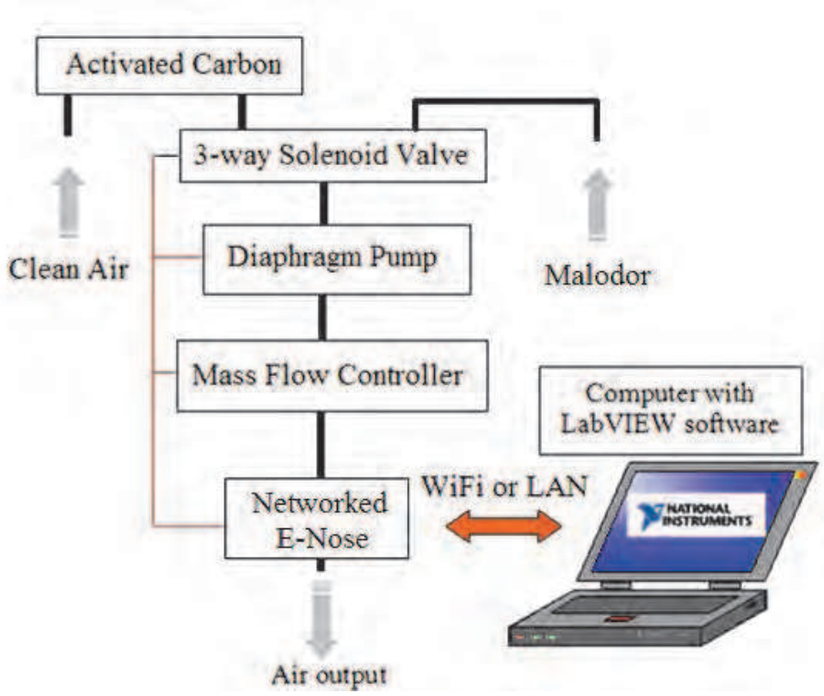

Fig. 15. Schematic diagram of air flow system

4.2.2 Detection system

The commercial MOS gas sensors widely known as TGS (Tagushi) gas sensors can be used for the sensing part. The resistance measurement is performed as shown in Fig. 16 by applying a constant excitation current of I_C=10μA to each sensor element and measure the voltage drop U_1 via V_1.

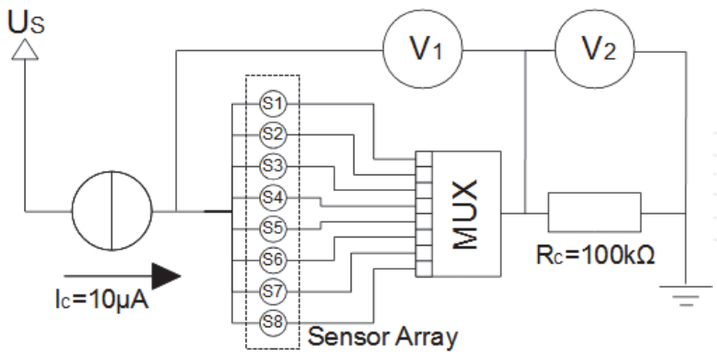

Fig. 16. Resistance measurement principle

The resistance can be calculated by the formula

$$R=U_1/I_C \tag{4.7}$$

An 8-Channel multiplexer was used to address each gas sensor. The voltage U_1 was measured between the input to all sensors and the output of the multiplexer (MUX). The

differential voltage U_1 was measured using the first channel of a 24-Bit Delta-Sigma ADC defined as V_1. The second channel measures the voltage drop U_2 defined as V_2 over a fixed resistor R_C=100kΩ that is in series with the multiplexers and the sensor matrix. Therefore, it is possible to measure the constant current and calculate corrections if the current changes due to temperature effects or external noise.

4.2.3 Control and data analysis system

From section 4.2.2, the ADC, MUX and excitation current source are part of a measurement system controlled by a Microchip PIC24HJ microcontroller. Additionally, a digital-analog converter (DAC) was used to set the flow rate of a mass flow controller and one ADC input of the microcontroller was used to measure the flow rate feedback. Also the 3-way solenoid valves were controlled by the microcontroller. The microcontroller is interfaced to an Ethernet controller, so the measurement system can make an Ethernet connection to a personal computer (PC) on board the measurement software written in LabView. By connecting the measurement system to a small WiFi-Router, this E-nose could become a flexible solution for use in a wireless sensor network. It is possible to use encrypted communications to the PC that can make the system to be secure. The PCB board that contains the metal oxide gas sensors and a temperature/humidity sensor was plugged onto the measurement system PCB board. This creates a flexible setup with interchangeable sensors. A photograph of a networked E-nose is displayed in Fig. 17.

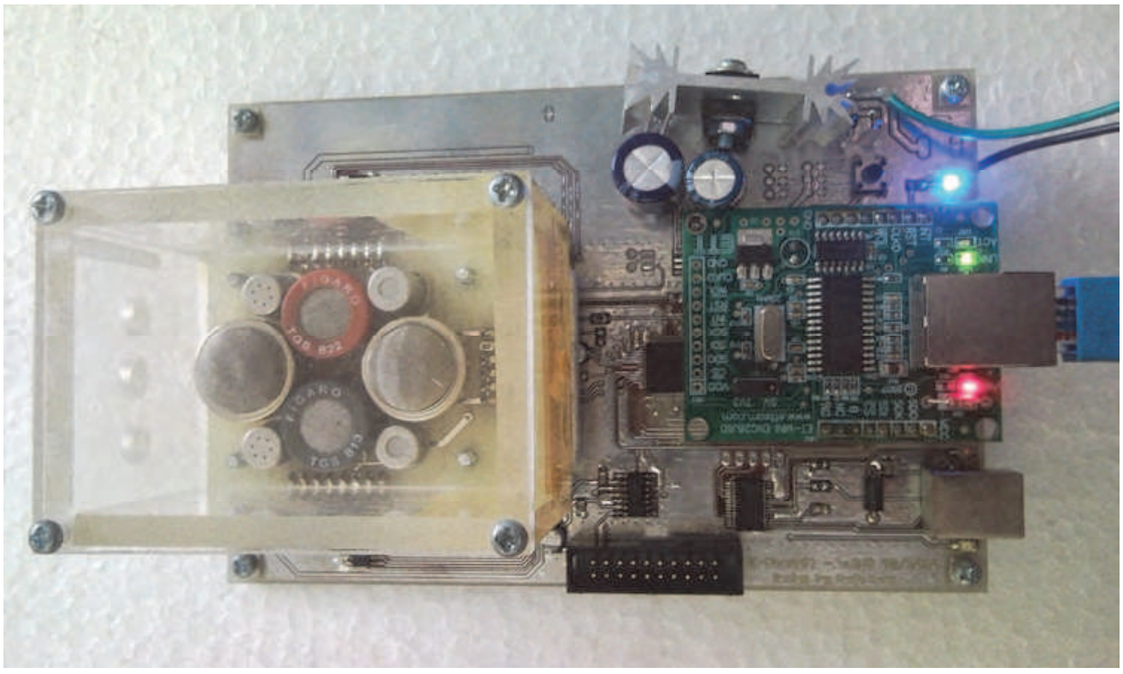

Fig. 17. Photograph of a networked E-nose (15 cm x 10 cm) developed by Center of Nanoscience and Nanotechnolohy, Faculty of Science, Mahidol University.

4.2.4 Other networked E-nose

A wireless E-nose based on IEEE 802.15.4 (ZigBee wireless network) has been developed for environment quality classification as shown in Fig. 18 (Pogfay et al., 2010).

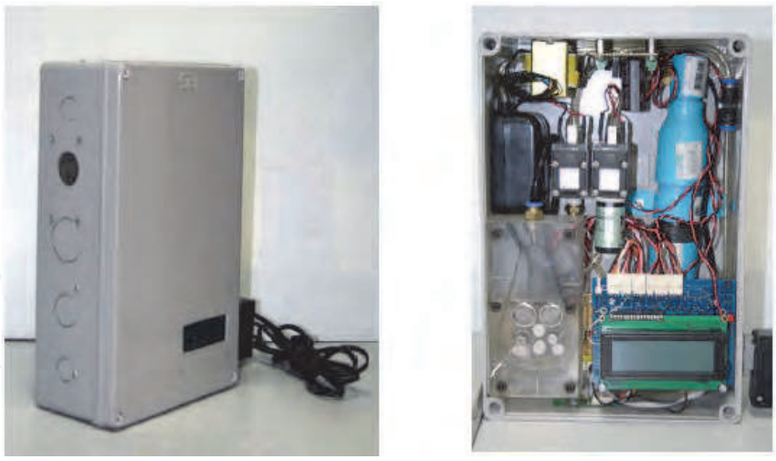

Fig. 18. Photographs of a wireless E-nose based on ZigBee wireless network (Pogfay et al., 2010).

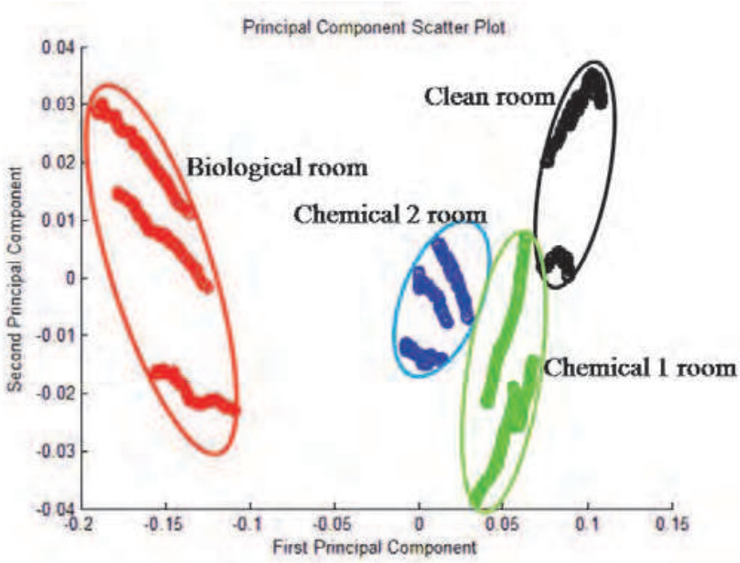

Fig. 19. 2-D PCA plot for environment quality classification (Pogfay et al., 2010).

The advantage of this ZigBee wireless network lies in its ability to offer low power consumption (50 mW) and extend the line of sight distance up to 1 mile. By combined with PCA analysis, the wireless E-nose can clearly classify the air environment from different rooms as shown in Fig. 19.

A smart wireless E-nose (Pan & Yang, 2009) has been designed to be compact in size, energy efficient, and low cost as shown in Fig. 20. This E-nose has been applied for online monitoring of livestock farm odors. Based on field applications like livestock farm monitoring, the circuit and environmental factor noises can strongly affect the output signals. To overcome the problems, a modified Kalman filtering technique has been developed for improving the sensor sensitivity and precision of odor strength measurement for livestock farm odors (Qu et al., 2009). The new odor strength measurement equation based on the noise analysis of MOS gas sensors can be modeled by (Qu et al., 2009):

$$y_k = Cx_k + d_k + s_k \tag{4.8}$$

where C is a measurement matrix, x_k is the system state vector, d_k is the direct current noise component with the same frequency as the signal and s_k is the white noise.

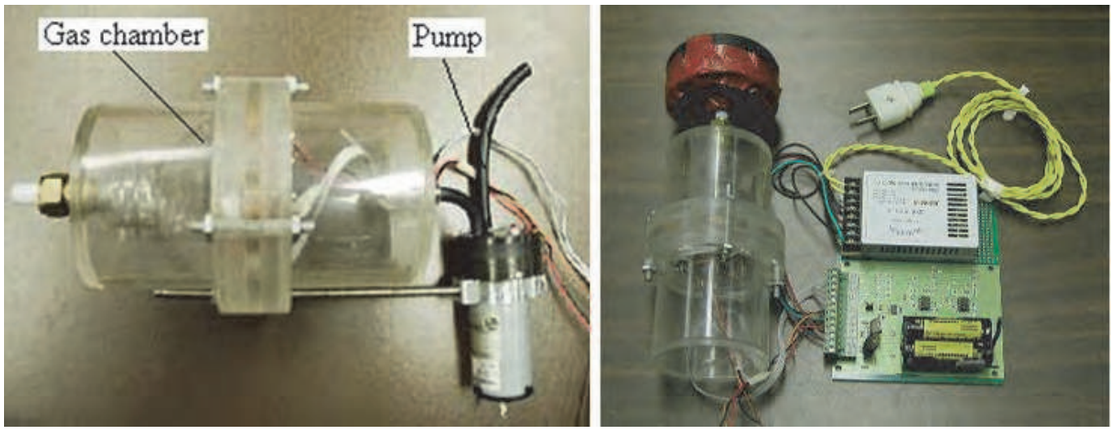

Fig. 20. The smart wireless E-nose (Qu et al., Pan & Yang, 2009).

5. Acknowledgements

Mahidol University and National Research Council of Thailand are acknowledged for supports.

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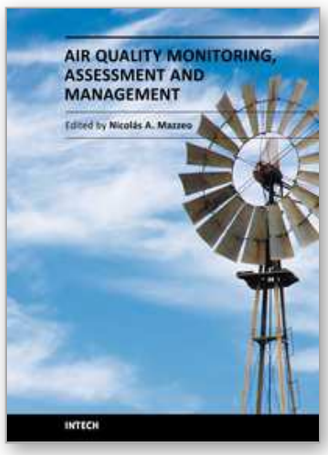

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Human beings need to breathe oxygen diluted in certain quantity of inert gas for living. In the atmosphere, there is a gas mixture of, mainly, oxygen and nitrogen, in appropriate proportions. However, the air also contains other gases, vapours and aerosols that humans incorporate when breathing and whose composition and concentration vary spatially. Some of these are physiologically inert. Air pollution has become a problem of major concern in the last few decades as it has caused negative effects on human health, nature and properties. This book presents the results of research studies carried out by international researchers in seventeen chapters which can be grouped into two main sections: a) air quality monitoring and b) air quality assessment and management, and serves as a source of material for all those involved in the field, whether as a student, scientific researcher, industrialist, consultant, or government agency with responsibility in this area.

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