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Review on MOS Gas Sensors based e-Noses and their Applications to Food Analysis

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ABSTRACT: An e-nose is an electronic instrument which consists of chemical sensors array having partial specificity and necessary pattern recognition tool, capable of recognizing different types of odors. This review focuses on commercially available metal oxide gas sensors and its use in commercial as well as experimental e-noses. The review covers fabrication and working of MOS based gas sensors, material used for fabrication, Processes, Companies that are producing various range of gas sensor modules for different gases, commercial e-nose systems using such sensors and typical application they can handle. The review also focuses on use of MOS based e-noses for quality control applications to food and beverages, determination of storage shelf-life or freshness, detection of contaminants or adulteration of wide range of foods products including: fish, meat, grains, alcoholic drinks, non-alcoholic drinks, fruits, milk and dairy products, fresh vegetables, olive oils, nuts, and eggs etc.

KEYWORDS: Electronic Nose, food analysis, gas sensors, MOS sensor array, odor sensing,Commercial e-noses

I.INTRODUCTION

E-noses consist of two main components that are sensing and pattern recognition system. The sensing system is consists an array of different sensing elements. VOC Vapour presented to the sensor array produces a signature pattern which is characteristic of the vapour. Database of vapour signatures can be produced by presenting different gas vapors to sensor array. Pattern recognition is fundamental part of any e-nose system. The PARC methods categorized in to three main classes depending upon available data and the type of result required. The graphical analysis with bar charts, Radar plots, offset polar plots and profile polar plots are simple form of data analysis technique used in e-noses. The second way of analysing data is multivariate analysis which reduces data dimensionality including techniques such as PCA, PLS, CLA, LDA etc. The third way is ANN (Artificial neural network), which consists of a set of interconnected processing algorithms functioning in parallel. ANN is based on the cognitive process of the human brain [1, 2].

Metal oxide semiconductor (MOS) based gas sensors are readily available commercially. They are most widely used type to make array for odor sensing than any other class of gas sensors [1].There are many metal oxides which shows gas sensitivity under suitable condition but tin dioxide (SnO_2) doped with a small amount of a catalytic metal such as palladium or platinum widely used. Various SnO2 based resistive sensors have been developed for a range of applications by deciding the proper catalyst and operating Conditions [2].Other MOS based gas sensors includes titanium dioxide (TiO_2), zinc oxide (ZnO), and tungsten oxide (WO3) etc [3].The MOS sensor performance is not only depends on the composition of sensor materials but also deposition is vital variable governing sensor performance design [4].There are various deposition techniques includes thermal evaporation, spraying, physical or chemical vapour deposition for thin films of 6–1,000 nm thickness and screen printing or painting for thick films of 10–300 μm thickness. Thin film devices have a faster response and having good sensitivities but manufacturing process is complex. So thick film technology oftenly used in commercially available MOS based gas sensors [2].

The MOS based gas sensor usually consists of a ceramic support tube contains a platinum heater coil and outside of support tube is coated with sintered SnO_2 material with any catalytic additives. The electrical resistance change occurs in MOS based sensors when exposed to odorant gases. Resistance changes occur due to combustion reactions with the lattice oxygen species on the surface of metal oxide material [1]. The metal oxide can be P type or N type. These types

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can be achieved by doping process. The resistance of MOS material increase or decrease depends on type of material. Resistance increases in P type and decreases in N type due to presence of oxidizing gases [3].SnO_2 and WO_3 are examples of n type; CTO is example of p-type metal oxide semiconductor. An e-nose has ability to estimate odors quickly though it has little or no alikeness to animal noses. Sensors used in E-noses requires independent and narrowly tuned to certain VOCs, compared with olfactory receptors from invertebrate animals like fruit flies [5].

II. MOS SENSOR BASICS

Metal oxide semiconductors have been used as a gas sensing element. The MOS is composed of a heating element coated with a semiconductor, most typically tin oxide (SnO_2) doped with small amounts of catalytic metal additives, for e.g. palladium, platinum etc. The selectivity to different odorants can be archived by doping process. Figaro Engineering is one of the major manufacturers of the MOS based sensor, whose sensors were developed by N.Taguchi [6], hence the name Taguchi Gas Sensor (TGS). The TGS sensor operates by allowing electrical current to flow through the grain boundaries of the SnO_2 micro-crystal surface. At the grain boundary, oxygen is adsorbed and forms a potential barrier.

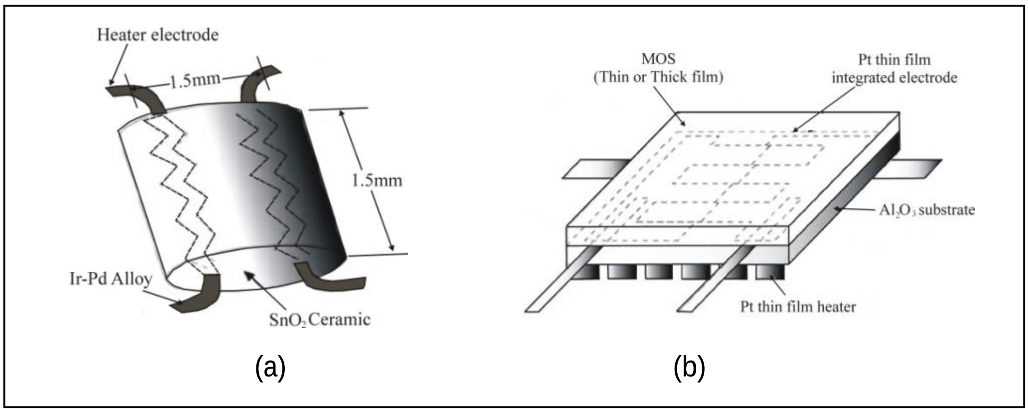

Fig.1Construction of commercially available MOS gas sensorsa) Sintering type b) Thin film type

The size of this potential barrier is reflected in the value of the sensors resistance, the higher the barrier more is the resistance across the sensor. Contrarily, exposure to a deoxidizing gas will deplete the grain boundaries and thus electrical charge flows more freely and the overall resistance is reduced.

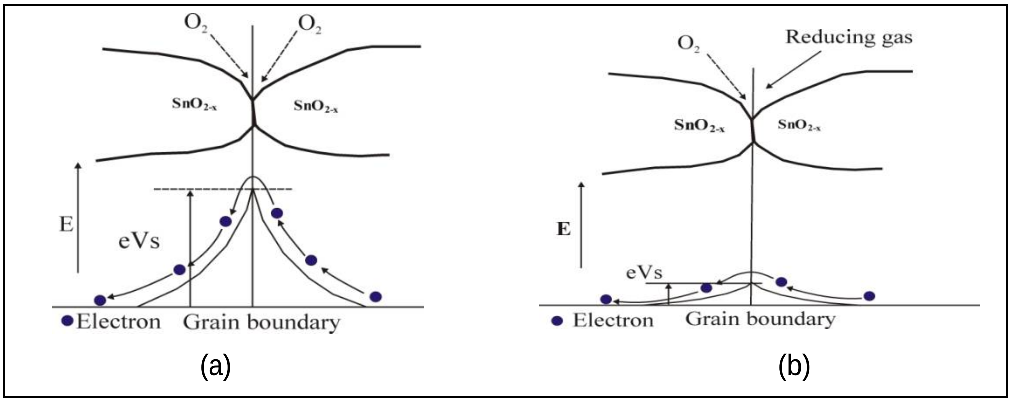

Fig. 2 :Schematic diagram explaining the conductivity status of sensor using energy band diagram : a) Potential barrier in the absence of reducing gas. b) Potential barrier in the presence of reducing gases

The metal oxide sensing layer can be thick or thin. Thin films have many advantageous properties including faster response and greater sensitivity to strongly oxidizing species, while thick films are more easily and reproducibly

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fabricated. Micro fabrication of MOS sensors allows multi sensors to be packed on a single chip [7], produced a 16.2 mm by 4.25 mm integrated array of six inter-digitated sensors using micromachining. Each sensor normally is sensitive to several deoxidizing gases, with most sensitivity to an optimized gas determined by the fabrication of the physical sensing materials. A final requirement to sensor operation is to supply an input power source for the internal heater. MOS based gas sensors operate at high temperature between 300^{o}C to 500^{o}C, that increases the sensitivity characteristics of it. Although, the MOS sensors are less selective, they have been used in the design of electronic nose systems, as they are small in size, low cost and easily available. The most attractive feature of these gas sensors is having sensitivity to wide range of gases, usable life span of 3 – 5 years depending on the usage of the sensor. A few drawbacks of the sensors however, include a dependency between external humidity and temperature effects to the rate of reaction to a gas and high power consumption. These drawbacks can be overcome by using appropriate signal pre-processing techniques before employing the pattern recognition algorithm.

III.COMMERCIALLY AVAILABLE MOS GAS SENSORS

Gas sensors based on MOS are commercially available. They have been more widely used to make e-nose arrays for odor measurement than any other class of gas sensors. Table 1 lists some of the commercially available gas sensors of SnO_2 and ZnO that are manufactured by Figaro Engineering Inc. (Japan), HANWEI ELETRONICS CO., LTD

Table 1: Application of MOS based E-noses in the food industry, sensors used and performance.

Manufacturer	Model	Target Gas Sensitivity
FIGARO ENG	TGS2600	Air contaminants (hydrogen, ethanol, etc.
	TGS2602	Air contaminants (VOCs, ammonia, H_2S, etc.)
	TGS2603	Air contaminants
	TGS8100	Air contaminants (hydrogen, ethanol, etc.)
	TGS826	Ammonia
	TGS2444	Ammonia
	TGS3870	Methane, Carbon Monoxide
	TGS821	Hydrogen
	TGS823	Alcohol, Solvent vapours
	TGS2620	Alcohol, Solvent vapours
	TGS823	Alcohol, Solvent vapours
	TGS2620	Alcohol, Solvent vapours
	TGS816	Methane, Butane, Propane
	TGS2610-00	Butane, Propane
	TGS2610-00	Butane, Propane
	TGS2612	Methane, Propane, Butane
	TGS6810-00	Methane, Propane, Butane
	TGS6812-00	Hydrogen, Methane, Butane, Propane
	TGS832-A00	Halocarbon gas
	TGS832-F01	Halocarbon gas
	TGS816	Methane, Butane, Propane
	TGS2611-00	Methane
	TGS2611-00	Methane
	TGS816	Methane, Butane, Propane
HANWEI LTD.	MQ-2	General combustible gas
	MQ-3	Alcohol Vapour
	MQ-4	Natural gas, Methane
	MQ-5	LPG, Natural gas, Coal gas
	MQ-6	LPG, Propane
	MQ-7	Carbon Monoxide (CO)
	MQ-8	Hydrogen
	MQ-9	CO and Combustible gas
	MQ131	Ozone O_3
	MQ135	Air Quality Control (NH_3,Benzene,Alcohol,smoke)
	MQ136	Sulphur dioxide SO_2
	MQ137	Ammonia (NH_3)
	MQ138	VOC (Mellow, Benzene, Aldehyde, Ketone, Ester)
	MQ216	Natural gas\Coal gas
	MQ303A	Alcohol
	MQ306A	LPG, Propane
	MQ309A	Carbon Monoxide (CO), Flammable Gas

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IV. MOS GAS SENSOR ARRAY BASED COMMERCIAL E-NOSE SYSTEMS

Commercially there are lots of manufacturers who produce a wide range of applications oriented e-nose products using MOS sensor technologies. Upto 2016 there were at least 9 major manufacturers selling electronic nose instruments of various types based on MOS technology and these companies are summarized in Table 2 together with application(s).

Table 2:Some commercial companies that manufactured e-noses based on MOS sensors upto 2016

Manufacturer	MODEL	No. of Sensors	Typical Applications	Ref.
Airsense Analysis GmbH	PEN3	10	Food evaluation, Flavour, Fragrance	http://www.airsense.com/
Alpha MOS-Multi Organoleptic Systems	FOX 2000,FOX3000, FOX4000,Gemini, RQ Box, Prometheus,	6-18	Quality control of food storages, Analysis of dairy products, Alcohol beverages and perfumes	http://www.alpha-mos.com
AltraSens	Odor Vector	6	Odor filters monitoring, Monitoring odor emissions from a sewage canal.	http://www.altrasens.com/
Dr. Fodisch	OMD 98, OMD 1.10	10-12	Odor pollutions from industry, agriculture and recycling of waste.	http://ankersmid.ro/
Environics, USA	ChemPro 100	3	chemical warfare agents and toxic industrial compounds/materials	http://www.environics.fi/
GERSTEL	QCS	n.d.	VOC emissions from beverage cans	http://www.gerstel.com/
RST-Rostock	FF2	6	Fire detection, propane and methane emissions	http://www.rst-rostock.com/
SACMI	EOS Ambiente, EOS835,EOS Aroma	6	VOC detection in landfills, urban solid waste selection and treatment plants, biogas production plants, composting plants , WDF production plants, incinerators and waste to energy plants, civil and industrial waste water treatment plants, livestock farming, rendering plants, distilleries, refineries ,chemical plants	http://www.sacmi.com
Sensigent	Cyranose 320	32	petrochemical, chemical, food and beverage, packaging materials, plastics, pet food, pulp and paper, medical research etc.	http://www.sensigent.com/

n.d- not disclosed

V. APPLICATION OF MOS BASED E-NOSES IN FOOD INDUSTRY

This review article focuses on the use of MOS gas sensor based electronic noses for food applications. The review also focused on the technical limitations for some applications and to solve this issue how the different ways is choose to overcome them. The MOS gas sensor based e-noses those are related to quality control, monitoring process, geographical origin, ageing, adulteration, ripening state, contamination and spoilage summarized in Table 3.

Table 3: MOS sensor based electronic noses for food applications

Food type	Test	E-nose type	No. of MOS sensors	Material	Ref
Fruit	Ripening changes	Commercial	10	SnO_2	[8]
		Experimental	2	SnO_2	[9]
		Commercial	10	SnO_2	[10]
		Experimental	5	SnO_2	[11-12]
		Commercial		SnO_2, CTO, WO_3	[13]
	Varieties	Experimental	16	SnO_2 thin film doped with Cr, Pt	[14-15]
	Microbial contamination	Commercial	6	SnO_2	[16]
	Shelf life	Commercial	10	SnO_2	[17]
		Commercial	18	SnO_2, CTO, WO_3	[18]
Meat	Freshness and type of meat	Commercial	4	SnO_2	[19]
	Microbiological	Experimental	7	SnO_2	[20]

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	contamination				
	Origin of meats	Commercial	6	SnO_2	[21]
	Taints	Experimental	5	SnO_2	[22]
Fish	Freshness	Commercial	6	SnO_2, CTO, WO_3	[23-24]
		Experimental	6	SnO_2	[25]
dairy products	Adulteration/ Contamination	Commercial	10	SnO_2	[26]
	Off-flavour	Commercial	12	SnO_2	[27]
	Cheese type	Commercial	6	SnO_2, CTO, WO_3	[28]
	Bacterial strains	Commercial	12	SnO_2, CTO, WO_3	[29]
	Origin of milk	Commercial	18	SnO_2, CTO, WO_3	[30]
Eggs	Freshness	Commercial	12	SnO_2 thick film	[31]
	Freshness	Experimental	4	SnO_2	[32]
Grains	Contamination by toxin	Commercial	6	SnO_2	[33]
	Off-flavors	Experimental	4	SnO_2	[34-35]
Olive oil	Authenticity	Commercial	6	SnO_2, CTO, WO_3	[36]
	Taints	Commercial	18	SnO_2, CTO, WO_3	[37]
Alcoholic beverages	Denomination of origin and vineyard discrimination	Experimental	4	WO_3	[38]
		Experimental	16	SnO_2 thin film doped with Cr, In	[39]
		Commercial	10	SnO_2	[40]
	Aging of wines and beers	Experimental	16	SnO_2 thin film doped with Cr, In	[41]
		Experimental	20	SnO_2	[42]
		Commercial	12	SnO_2, CTO, WO_3	[43]
	Classification of drinks	Commercial	12	SnO_2, CTO, WO_3	[44]
		Commercial	6	SnO_2, CTO, WO_3	[45]
		Experimental	6	SnO_2	[46]
		Commercial	6	SnO_2	[47]
	Off-flavours and aromatic compounds	Commercial	12	SnO_2, CTO, WO_3	[48]
		Experimental	16	SnO_2	[49]
Non alcoholic beverages	Grading	Experimental	4	SnO_2	[50]
		Commercial	10	SnO_2	[51]
	Quality/process control	Experimental	6	SnO_2	[52]
		Experimental	12	SnO_2	[53]
		Experimental	4	SnO_2 thin film doped with Au, Pt	[54]
		Commercial	6	WO_3, SnO_2	[55]
Other food	Shelf life of nuts	Commercial	10	SnO_2	[56]
	Shelf life of vegetables	Experimental	5	SnO_2	[57]
	Bacterial species classification	Commercial	6	SnO_2, CTO, WO_3	[58]

VI. CASE STUDY

The Guava fruit classification based on its ripening state at the time of harvest was done using indigenously developed e-nose based on MOS gas sensor array details given in [59] and the data acquired from it was analysed using radar plot technique.

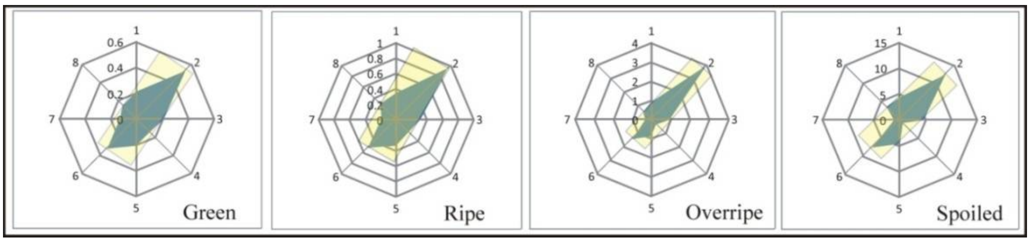

Fig. 3 :Radar plots explaining the descriminating power of developed e-nose for green,ripe,overripe and spoiled class.

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The different odor signature was observed from the analysis. The bounding box as well as aspect ratio of the odorprint was calculated and compared for four different ripening states of guava fruits as green, ripe, overripe and spoiled shown in table 4. This indicates the use of non-specific selective gas sensor arrays to construct an odor database.

Table 4: Observations for different ripening state of guava fruit using e-nose

Fruit Class	Bounding box area	Aspect ratio
Green	0.168	4.2
Ripe	0.568	3.55
Overripe	5.94	4.9
Spoiled	133	2.71

In the fig 4, it shows the graph of ripening state og guava fruitVsbounding box area of odorprint shown in radar plot.

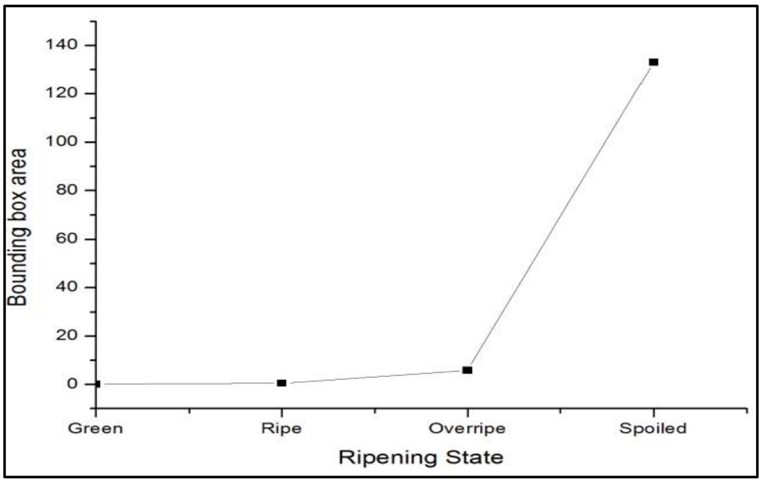

Fig. 4: Ripening state of fruit v/s bounding box area.

From figure 4 it is clear that as ripening state increases from green to spoiled; the bounding box area also increases, which indicates increase the VOC emission and change in aspect ratio indicates gas content in VOC changes with ripening.

VII. CONCLUSION

A variety of sensing technologies, different in their principle of operations, has been utilized for e- noses and each type has been applied to range of problems. Sensors used in e-noses must have small in size, inexpensive, robust and semi-selective. The proper selection of sensors for the e-noses may be useful for particular applications. The MOS based sensors attains these requirements and are most widely used because of low cost and small size. The potential use of such sensors in various types of e-noses were reviewed for food area; including applications in quality control, determination of geographical origin ,monitoring process, aging, contamination and spoilage, adulteration etc. The sensor materials and designs, limitation of currently available MOS sensors with their independence and selectivity is being reported. The case study is reported for indigenously developed e-nose.

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