
Porous Silicon Gas Sensing

Giuseppe Barillaro

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Abstract

In this chapter, the state of the art on porous silicon gas sensors, both electrical and optical, is reviewed by paying special emphasis on the advancement of gas sensor architectures that has occurred over the two last decades, as well as on the different functionalization approaches implemented in and chemical species sensed with such architectures. Ten main architectures, five for the electrical domain (capacitor, Schottky-like diode, resistor, FET-like transistor, and junction-like diode) and five for the optical domain (single layer, waveguide, Bragg mirror, resonant cavity, and rugate filter), have been proposed so far for improving gas sensor features. Several functionalization schemes have been integrated in such architectures to improve sensor performance, and more than 50 different chemical species have been sensed using porous silicon gas sensors. The latest trends on multiparametric sensing on single devices as well as on multisensor integration in a single chip, for both optical and electrical domains, are also discussed.

G. Barillaro (✉)

Dipartimento di Ingegneria dell'Informazione, Università di Pisa, Pisa, Italy

e-mail: g.barillaro@iet.unipi.it

Introduction

Gas sensors, which are a subset of the broader family of chemical sensors, allow inferring on the chemical species present in the surrounding environment. An ideal gas sensor should have high sensitivity, high specificity, small limit of detection, high resolution, high accuracy, high precision, large dynamic range, null offset, high linearity, null hysteresis, short response time, and long operation life (see Korotcenkov (2013)). Of course, no current gas sensors meet all these requirements simultaneously, which are, on the other hand, neither achievable nor necessary at the same time in real-world applications.

The analysis of the state of the art of porous silicon gas sensors highlights how the early studies were mostly focused on the development of suitable readout approaches with the aim of investigating the sensing properties of porous silicon layers in sensor structures featuring both basic architecture (e.g., capacitor, monolayer) and simple surface chemistry (e.g., native surface or oxidized surface). On the other hand, the latest studies are mostly focused on the integration of porous silicon layers in sensor structures featuring advanced architectures (e.g., FET-like transistors, stacked rugate filters, etc.) and sophisticated surface chemistry (e.g., silanization, carbonization, metallization, etc.) with the aim of improving sensitivity, selectivity, and reliability performance. Finally, optical and electrical sensing platforms integrating array of porous silicon sensors with advanced features on the same chip are envisaged in the near future, though still in their infancy today (see handbook chapter “► Porous Silicon for Microdevices and Microsystems”) (Fig. 1).

Gas Sensing with Porous Silicon

In the last two decades, micro-, meso-, and nanostructured forms of silicon, namely, porous silicon, have been demonstrated to be very effective for the fabrication of integrated gas sensors with low-cost process and room temperature operation (see Mizsei (2007), Ozdemir and Gole (2007), Saha (2008)). The increased specific surface (by definition, accessible surface to volume ratio) of porous silicon, up to 10^7 times larger than bulk materials, ensures a stronger interaction between material

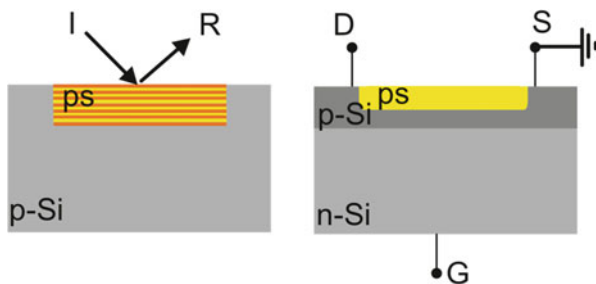


Fig. 1 Sketch of a multilayered optical device (on the *left*) and a FET-like electrical device (on the *right*) representative of the last-generation architecture of porous silicon gas sensors

surface and gas molecules and allows high sensitivity and good limit of detection to be achieved for a large number of gaseous species (e.g., NO_x and other inorganic gases, organic compounds, among which explosives, hydrocarbons, alcohols, halides, amines, ketones, etc.). Optical, electrical, and electrochemical approaches have been established to be valuable for the detection of a multitude of different gas species (both inorganic and organic). Changes in optical (e.g., refractive index, radiative recombination processes, etc.) and electrical (e.g., dielectric constant, conductivity, etc.) properties of the porous silicon material upon interaction (adsorption and/or condensation processes) with the specific gas species have been demonstrated through quantitative monitoring of the variation of different parameters (e.g., photoluminescence spectrum; reflected, transmitted, and diffracted optical power; capacitance; current; resistance; etc.) as a function of the gas concentration. A number of architectures have been proposed for both electrical (e.g., metal-based devices, among which capacitor, resistor, and Schottky diode, and pn junction-based devices, among which diode, transistor, etc.) and optical (e.g., monolayer-based devices, among which waveguide, and multilayer-based devices among which Bragg mirror, resonant cavity, rugate filter, etc.) gas sensors with the aim of improving sensor performance (e.g., improve sensitivity, limit of detection and selectivity, compensate for baseline drift, compensate for measurement angles, etc.). An equivalent large number of readout approaches (e.g., spectrometry, interferometry, ellipsometry, birefringence, conductometry, impedance spectroscopy, etc.) have been reported for monitoring the sensor parameters, by using both single-parameter and multiparameter approaches. Besides, a number of functionalization schemes (e.g., oxidation, hydrosilylation, carbonization, metallization, etc.) of the porous silicon surface have been proposed with the aim of reducing aging and interfering effects while improving reliability, sensitivity, and selectivity. See, for example, the handbook chapter “► [Silicon-Carbon Bond Formation on Porous Silicon.](#)” Tables 1 and 2 provide the main architectures of porous silicon electrical and optical gas sensors, respectively, since 1990. The porous silicon physical parameters that mainly account for the variation of the sensed quantity for the specific sensor architecture are also indicated along with the gas species sensed (Table 3).

Sensing Platforms

More recently, in addition to mere sensor devices exploiting either single-parameter or multiparameter monitoring, miniaturized sensing platforms integrating a number of porous silicon optical sensors featuring different surface chemistry on the same chip, as well an array of porous silicon electrical sensors together with CMOS electronic driving/readout circuits on the same chip, have been demonstrated to be feasible, thus envisaging the realization of a new class of electronic and photonic system-on-chips with gas sensing capability.

Table 4 provides study examples on up-to-date sensing platforms exploiting porous silicon either electrical or optical gas sensors.

Table 1 Main architectures of porous silicon electrical gas sensors since 1990

Electrical Sensors		Metal-based devices			Junction-based devices		
Sensor architecture	Capacitor	Schottky-like diode	Resistor		FET-like transistor	Junction-like diode	
Physical parameter	Dielectric constant	Energy barrier	Conductivity	Conductivity	Conductivity	Energy barrier	
Sensed quantity	Capacitance	Conductivity	Resistance	Resistance	Dielectric constant	Conductivity	
Sensed species	Inorganic gases	Current	Inorganic gases	Inorganic gases	Current	Current	
	Inorganic gases	Inorganic gases	Inorganic gases	Inorganic gases	Impedance		Inorganic gases
	Aliphatic organic substances	Aliphatic and aromatic organic substances	Aliphatic and aromatic organic substances	Aliphatic and aromatic organic substances	Inorganic gases		Inorganic gases
Study examples	(Andersson et al. 1990; Ben-Chorin and Kux 1994; Rittersma et al. 2000; Foucaran et al. 2000; Belhousse et al. 2004; Björkqvist et al. 2004a, b, 2009; Gabouze et al. 2005)	(Ben-Chorin et al. 1994; Schechter and Ben-Chorin 1995; Mareš et al. 1995; Stievenard and Deresnes 1995; Bilenko et al. 1997; Foucaran et al. 1997; Zhang et al. 2000; Pancheri et al. 2003; Gabouze et al. 2004a, 2006; Razi et al. 2010; Kanungo et al. 2010)	(Gabouze et al. 2005; Björkqvist et al. 2009; Motahashi et al. 1995; Talliercio et al. 1995; Angelucci et al. 1999, 2000; Mulloni et al. 2000; Di Francia et al. 2000; Baratto et al. 2000, 2001; Boarino et al. 2000; Šetkus et al. 2001; Seals et al. 2002; Oton et al. 2003; Chakane et al. 2003; Galeazzo et al. 2003; Massera et al. 2004; Amato et al. 2004; Gaburro et al. 2004b; Lewis et al. 2005, 2007; Luongo et al. 2005; Rahimi and Irajizad 2007; Sankara Subramanian et al. 2007; Gole et al. 2007; Galstyan et al. 2008; Kovacs and Meister 2009; Peng et al. 2009, 2012; Ozdemir and Gole 2010; Moshnikov et al. 2012; Laminack and Gole 2013)	(Barillaro et al. 2003, 2004, 2005; Archera et al. 2005; Barillaro et al. 2006, 2010a, b; Lazzarini et al. 2013)			(Barillaro et al. 2007a, 2008; Prabakaran et al. 2008)
Timeline	Chronological order of exploitation of the different architectures for gas sensing since 1990						

Table 2 Main architectures of porous silicon optical gas sensors since 1990

Optical sensors		Multilayer-based devices			
Sensor architecture	Monolayer-based device		Bragg mirror		Rugate filter
	Single layer	Waveguide	Resonant cavity	Refractive index	Refractive index
Physical parameter	Radiative recombination efficiency	Refractive index	Refractive index	Refractive index	Refractive index
Sensed quantity	Reflectance spectrum/intensity	Transmission losses	Resonance peak position	Resonance peak position	Resonance peak position
Sensed species	Inorganic gases	Aliphatic organic compounds	Aliphatic and aromatic organic substances	Aliphatic and aromatic organic substances	Inorganic gases
Study examples	Aliphatic and aromatic organic substances	Aliphatic organic compounds	Aliphatic and aromatic organic substances	Aliphatic and aromatic organic substances	Aliphatic and aromatic organic substances
	(Ben-Chorin et al. 1994; Baratto et al. 2000; Lauerhaas et al. 1992; Lauerhaas and Sailor 1993; Coffier et al. 1993; Lee et al. 1995; Rehm et al. 1995; Rehm et al. 1996; Harper and Sailor 1996; Kelly et al. 1996; Song and Sailor 1997; Kelly and Bocarsly 1998; Content et al. 2000; Holec et al. 2002; Chvojka et al. 2004; Salcedo et al. 2004; Dian et al. 2005, 2010; Vrkoslav et al. 2006, 2007; Jelínek et al. 2007; Bjorklund et al. 1996; Zangoie et al. 1997; Gao et al. 2000, 2002a, b; Letant and Sailor 2000; Létant et al. 2000; Wang and Arwin 2002; Lita et al. 2002; Lin et al. 2004; Geobaldo et al. 2004; Torres-Costa et al. 2009)	(Arrand et al. 1999; Rea et al. 2009)	(Zangoie et al. 1998; Snow et al. 1999; Allcock and Snow 2001; Torres-Costa et al. 2005; Moretti et al. 2007; Descrovi et al. 2007; Kim et al. 2010, 2011; Jalkanen et al. 2010)	(Mulloni et al. 2000; Zangoie et al. 1999; De Stefano et al. 2003, 2004a, b)	(Li et al. 2003; King et al. 2007; Ruminski et al. 2008, 2010; Chapron et al. 2007; Shang et al. 2011a; Kelly et al. 2011a, b)
Timeline	Chronological order of exploitation of the different architectures for gas sensing since 1990				

Table 3 Sensed species with electrical and optical porous silicon gas sensor architectures of Tables 1 and 2

Substances sensed with electrical and optical porous silicon gas sensors	
Inorganic gases	Hydrogen, oxygen, water, carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO ₂), halogens (F ₂ , Cl ₂ , Br ₂ , I ₂), hydrofluoric acid (HF), hydrochloric acid (HCl), ammonia (NH ₃), hydrogen sulfide (H ₂ S), sulfur dioxide (SO ₂), phosphine (PH ₃)
Aliphatic organic substances	Methane, ethane, propane, butane, pentane, hexane, heptane, liquified petroleum gas (LPG), cyclohexane, ethylene, methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 1-pentanol, 1-hexanol, glycerol (1,2,3-propanetriol), diethyl ether, tetrahydrofuran, acetone, metylethylketone (2-butanone), n-propylamine, n-butylamine, n-pentylamine, triethylamine, tri-n-propylamine, ethyl acetate, acetonitrile, dimethylformamide, chloroform (trichloromethane), methylene chloride (dichloromethane), trichloroethylene (C ₂ HCl ₃), cyclohexyltrichloroethylene
Aromatic organic substances	Benzene, toluene, chlorobenzene, nitrobenzene, 1,4-dinitrobenzene, dinitrotoluene (DNT), trinitrotoluene (TNT)

Table 4 Electrical and optical sensing platforms with porous silicon

Porous silicon sensing platforms				Study examples
Electrical	Array of FET-like transistor sensors	On-chip electronics	NO ₂ detection	(Barillaro et al. 2007b; Barillaro and Strambini 2008)
Optical	Array of rugate filter sensors	Different surface chemistries on chip	HF, HCl, aliphatic organic substance detection	(Ruminski et al. 2011; Shang et al. 2011b; Sweetman and Voelcker 2012)

Summary

Over the last two decades, electrical and optical gas sensors based on porous silicon have been tremendously improved, in terms of architectures, performance, and sensed species. On the one hand, single devices with high sensitivity, low limit of detection, and good selectivity have been achieved for different analytes, although reliability and lifetime still remain among the major challenges for both electrical and optical sensors. On the other hand, multiparametric sensing on single devices and multisensor integration in a single chip have been very recently reported for both optical and electrical approaches, thus pushing porous silicon gas sensors to a new generation of miniaturized sensing platforms. As to the latter, tremendous improvement due to simultaneous integration of sensors with electronic and photonic silicon circuits is expected for both approaches, respectively, in the next future.

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