Temperature sensor made of polymer-derived ceramics for high-temperature applications

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1. Introduction

Sensors that can be operated in harsh environments are highly desired for applications in a variety of high-temperature systems, such as turbine engines, coal gasification systems, and material processing systems. It is expected that these sensors can measure real-time operating conditions of the systems to provide information for feedback control and system optimization to increase efficiency and reduce pollution, as well as to monitor the health of structural components to improve safety. However, developing such kinds of sensors is not trivial. The critical challenge is that they must survive harsh environments of the systems, including high temperature, high pressure and severe oxidation/corrosion.

Up to now, several materials have been explored for microsensor applications. Among them, semiconducting silicon is the most studied material for microsensors due to its controllable electric properties and well-developed microfabrication techniques. However, Si-based sensors can not be used at temperatures higher than 350 °C since severe material degradation at elevated temperatures [1,2]. Silicon carbide is another widely used material for sensors and has been shown to provide better performance than silicon-based sensors in terms of high-temperature capability. Commercially available SiC-based sensors can be used up to 500 °C [3–5].

Recently, polymer-derived ceramics (PDCs) have been considered as promising materials for high-temperature sensor applications [6]. PDCs are a new class of high-temperature ceramics synthesized by thermal decomposition of polymeric precursors. Previous studies have revealed that PDCs possess a set of excellent structural and functional properties, including excellent high-temperature stability [7], high creep resistance [8,9], outstanding oxidation/corrosion resistance [10–14], high-temperature semiconducting behavior [15,16], and anomalously high piezoresistivity [17]. The direct chemical-to-ceramic processing of PDCs is compatible with many manufacturing techniques for making micrometer/nano-sized ceramic parts from the materials [18–21]. In addition, compared to other existing micro-fabrication technologies, the authors have demonstrated that such kind of material has good micro-mechanical machinability with feature size as small as 20 μm, and has been applied for both temperature and pressure measurements. This paper describes the use of polymer-derived SiAlCN (silicoaluminum carbonitride) ceramics (PDC) to fabricate a temperature sensor for high-temperature applications. A unique sensor head was designed and fabricated with Pt wires seamlessly embedded as electrodes. Material characterization test demonstrates that the resistance of the sensor head decreases monotonically with surrounding temperature, suggesting its readiness to be used for temperature measurement. In actual experiment (temperature up to 830 °C), the measurement of the PDC sensor demonstrates good repeatability to both unidirectional and bidirectional temperature variations for the total span of 10 h, and its measurement follows closely with the thermal couple measurement. These results demonstrated that the temperature sensors made of polymer-derived ceramics (PDC) have excellent accuracy and repeatability, and can be used in high temperature environment.

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Fig. 1. (a) Optical image showing SiAlCN sensor and Pt leads; (b) SEM image showing the interface between SiAlCN and Pt wire.

Fig. 2. The sensor resistance with respect to temperature max of 620 °C (inset: relationship in higher part of the temperature range); (b) natural logarithm relationship.

Fig. 3. (a) The resistance of the SiAlCN sensor head as a function of temperature; (b) A plot of resistance of the sensor head vs. temperature in a format of In(RP) vs. 1/T; the blue points is experimental result and the red solid line are computed from Eq. (1). (For interpretation of the references to color in figure legend, the reader is referred to the web version of the article.)

measurement [26,27]. Previous work has demonstrated the temperature dependence of the electric resistances for such material, suggesting its capability to be used for temperature measurement and heat flux measurement in high temperature and harsh environment [30,31]; however, actual sensors made of the materials have not been reported yet.

In this paper, we report a temperature sensor made of polymer-derived SiAlCN (silicoaluminum carbonitride) ceramic. The ceramic sensor probe was treated as a temperature-dependent potentiometer to simplify the design of the overall sensing system. The temperature can be obtained from the voltage over a shunt resistor connected in series to the probe. In the following sections, we will first present the fabrication and characterization of the ceramic sensor probe followed by the design of the entire sensing system. Finally, we will illustrate the testing results of the sensor performance.
2. Fabrication and characterization of the ceramic sensor head

In this study, the sensor head was made of polymer-derived SiAlCN ceramic by using commercially available polysilazane (HTT1800, Kion) and Aluminum-tri-sec-butoxide (ASB, Sigma–Aldrich) as the precursors, as well as dicumyl peroxide (DP, Sigma–Aldrich) as the thermal initiator [22]. Polymer-derived ceramics (PDCs) are a new class of high-temperature multifunctional ceramics synthesized by thermal decomposition of polymeric precursors [28]. The synthesis processing includes the following basic steps [28,29]: (i) synthesize/modify polymer precursors, (ii) shape and cross-link to form infusible polymer components/devices, and (iii) convert the polymer components/devices to ceramic ones by pyrolysis. The materials formed by this process are predominantly amorphous ceramic alloys, consisting of silicon, oxygen, carbon and nitrogen [29]. Other elements such as boron and aluminum can also be incorporated into the network by modifying the precursors to tailor and improve their properties [7,13].

During material preparation, firstly, 8.8 g of HTT1800 was mixed with 1.0 g of ASB at 120 °C for 24 h under constant magnetic stirring. After cooling down to room temperature, 0.2 g of DP was then added into the mixture under sonication for 30 min for completely dissolving DP. The obtained viscous liquid was solidified by heat-treatment at 150 °C for 24 h. The solid was then milled into fine powder of ~1 μm using high-energy ball milling. The powder was pressed into discs of 12.5 mm in diameter and 4–5 mm thick. Rectangular-shaped sensor heads were cut from the discs. In order to make electrodes, two holes of 350 μm diameter and 2 mm deep were drilled on the sensor head. The holes were then infiltrated with the liquid SiAlCN precursor; and Pt wires were inset into the holes. Finally, the assembled SiAlCN sensor head was pyrolyzed at 1000 °C for 4 h. The entire fabrication was carried out in high-purity nitrogen to avoid oxygen contamination.

Fig. 1a shows an optical image of the synthesized sensor head with dimensions of 3.4 mm × 2.7 mm × 1.2 mm. Fig. 1b is a SEM image showing the interface between the SiAlCN and Pt wires, revealing that they are well bonded together. I–V curve measurement suggested that a typical ohmic contact is formed between the SiAlCN and Pt wire.

In order to test the feasibility of using the SiAlCN for temperature sensing, the resistance of the fabricated sensor head was...
characterized by measuring its I–V curve using KEITHLEY 2400 (Keithley Instruments). Fig. 2 shows the material’s temperature dependent resistivity from 100 to 620 °C, and Fig. 3 is the relationship in the temperature range of 650–845 °C for a different batch of PDC sensor, where the temperature range is the focused range for this paper. Fig. 3a shows the resistance of the sensor head as a function of temperature. It is seen that the resistance of the sensor head monotonically decreases with temperature, suggesting that it can be used for temperature sensing.

The temperature-dependent resistance of the sensor head was further analyzed using Thermistor equation [23],

\[
\ln \frac{1}{R} = c_1 \frac{1}{T} - c_2
\]

where \(c_1\) and \(c_2\) are constants. To simplify the calculation, all resistances are in kiloohm (kΩ). Fig. 3b is a re-plot of the data in Fig. 3a in the format of \(\ln(1/R)\) vs. \(1/T\). The linear behavior indicates that the sensor head follows thermistor equation very well within our temperature range. The constants \(c_1\) and \(c_2\) are obtained by curve fitting to be \(-3235\) and \(-3.6\), respectively. The value of \(c_1\), which indicates the sensitivity of the material to the change of temperature, is comparable to other high temperature sensing materials. For example, for RF-sputtered SiC, \(c_1 = -3400\) [24]; and for single crystal SiC, \(c_1 = -1837\) [25].

3. Sensor design and characterization

The sensor head was simply modeled as a temperature dependent potentiometer and in order to comply with most commercially available data acquisition unit, the sensor resistance was converted to voltage using a simple circuit (Fig. 4a). The output voltage \(v_s\) can then be related to the sensor head resistance \(R_p\), the resistance of the shunt resistor \(R_s\), and input voltage \(v_i\) by

\[
v_s = \frac{R_s}{R_s + R_p} v_i
\]

Fig. 5. A plot of output voltage vs. temperature.

Fig. 6. PDC sensor response emulating thermocouple temperature to 900 °C.
In this study, a metal film resistor of 10 kΩ was selected as the shunt resistor.

The experimental setup is illustrated in Fig. 4b and the system block diagram including the associated data acquisition system is shown in Fig. 4c. The temperature sensing suite was characterized using a data acquisition (DAQ) system, which includes a real-time controller (PXI-8108, National Instrument, Austin, TX), an analog input/output DAQ card (PXI-6221, National instrument, Austin, TX), and a signal terminal box for flying leads connection (SCB-68 conditioning block, National Instrument, Austin, TX).

During the test, sensor probe was placed into high temperature regions of the tube furnace while the reference resistor was placed at room temperature. A K-type thermal couple was placed next to the PDC sensor head to provide a reference temperature, denoted as T. PXI-6221 can simultaneously record both $v_s$ and T. The input voltage is also provided by PXI-6221, which eliminates the need for an external power supply unit. The test was carried out in the range of 650–845 °C.

4. Results and discussion

4.1. Sensor parameters characterization

Fig. 5 shows the output voltage as a function of temperature measured using the set up shown in Fig. 4b. It is seen that the output voltage monotonously increases with increasing temperature. Such monotonous relationship between the voltage and the temperature satisfies the basic requirement for the sensing capability of the sensor. Fig. 6 is a set of our experimental result which compares the PDC sensor measurement with respect to the temperature output from the thermal couple. Overall the PDC follows the temperature trend very fluently with the exception of some discernible variance in the form of noise.

4.2. Repeatability test

Up to now the sensing capability of the designed system has been demonstrated by showing the monotonic corresponds between the output voltage and temperature. We now characterize the repeatability of the system, which is particularly important because it is fundamentally related to reliable measurements.

For doing so, the sensor was tested during heating and cooling cycles between 650 and 830 °C. First, the furnace was programed to increase the temperature from 650 to 830 °C then go back to 650 °C. The cycle contains several distinct steps with each step representing 25 °C (Fig. 7a). The sensor was tested for 6 cycles with the total span of 10 h. By doing such cycling test, both unidirectional and bidirectional repeatability can be characterized.

Fig. 7b shows the experiment results of the 6 cycles. Note that the different color lines represent the temperature measurements in different heating-cooling cycles. It is clear that the sensor produced almost identical and symmetric responses, which suggests very good repeatability performance.

4.3. PDC sensor for actual temperature measurement

Up to this point we have demonstrated the designed sensor by showing its measuring capability and good repeatability. We can now use the sensor to measure temperatures. The tube furnace in the experimental set up in Fig. 4b was programmed to periodically change its temperature between 680 and 730 °C. The temperature was measured by thermal couple and the sensor separately. Fig. 8c shows the measurement result from the thermal couple, which is consistent with the preset program. The direct measurement from the PDC sensor (Fig. 8a) is in voltage. In order to compare the results from thermal couple and the sensor, we need to convert the voltage output to temperature output. The temperature obtained from the PDC sensor is denoted as $T_{s}\text{sensor}$ which can be related to $v_s$ by combining Eqs. (1) and (2):

$$T_{s}\text{sensor} = \frac{c_1}{\ln\left(\frac{1}{v_s/v_t} - R_s\right) + c_2}$$ \hspace{1cm} (3)

Fig. 8b shows the results computed from Eq. (3) by using data in Fig. 8a. It is clear that the sensor system is capable of accurate temperature measurement; the maximum measurement error is about 4 °C at 730 °C. The major source of the error is believed to come from coefficients $c_1$ and $c_2$ being fitted over the range from 650 to 830 °C. The sensing accuracy can be further improved by using piecewise linear fit or a higher order fit.
5. Conclusion

A temperature sensor was successfully fabricated using polymer-derived SiAlCN ceramics, which is a new class of high-temperature multifunctional materials. The sensor head was first made from SiAlCN and characterized by measuring its temperature-dependent resistance. The result showed that the resistance of the sensor head monotonically decreases with increasing temperature, indicating its capability to measure temperature. A sensor was then designed by using a shunt resistor to convert resistance variation to voltage change for compatibility with commercially available DAQ devices. The parameters and repeatability of the sensor have been tested and validated. Finally, the sensor has been successfully implemented for temperature measurement and the result was compared to that from commercial thermal couple measurement.

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References


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Chengying Xu received the Ph.D. in 2006 in mechanical engineering from Purdue University, USA, and the M.S. in 2001 in mechanical manufacturing and automation from Beijing University of Aeronautics and Astronautics, China. She is currently an associate professor at the Florida State University, Tallahassee, Florida. Her research interests include high temperature sensor design, intelligent systems and control theory, manufacturing of advanced materials. Dr. Xu has co-authored a textbook: "Intelligent Systems: Modeling, Optimization and Control" (CRC Press, 2008, 433 pages) and four book chapters. She has authored and co-authored more than 30 journal papers and 30 refereed conference proceedings. She has served as an organizing committee member and session co-chair for a number of national and international conferences. She also works as an organizer for the Symposium Sensor Technology for the International Congress on Ceramics in Japan. She served as the Guest Editor for Transactions of the ASME, Journal of Micro- and Nano-Manufacturing, and has been an Associate Editor of the International Journal of Nanomanufacturing since 2008, and has been on the Board of Editor of Journal of Aviation and Aerospace Industry Manufacturing since 2010 and International Journal of Computational Materials Science and Surface Engineering since 2007.