Use of Polynomial Regression Analysis for Interpreting NO_x Sensitivity Response

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Abstract

The porous microstructure of Yttria-stabilized ZrO_2 (YSZ) affects the sensing capabilities of NO_x gas sensors. A Statistical method using R computing software was used to determine optimal conditions for the porous YSZ electrolyte microstructure. The electrolyte porosity is dependent upon fabrication firing temperature. Data indicated that the ideal fabrication temperature was 1041°C. Samples fired at 1041°C underwent impedance spectroscopy testing as well as porosity calculations to confirm the porous microstructure made had the expected effect on the sensing capability.

Regression Analysis

The statistical software R (r-project.org) was utilized in order to find the ideal fabrication firing temperature for NO_x sensors that would produce the highest sensitivity. However, using regression analysis for Sensitivity vs. Temperature yielded a low R-squared value. A polynomial model was then generated using θ, the phase angle response from the impedance data where the Δθ value is the numerator in the sensitivity equation: $\Delta\theta/\theta = (\theta_{x})/\theta_{O_2}$

```
Call:
lm(formula = D ~ T + I(T^2), data = data1)
Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  5.676e+01  1.526e+01   3.720   0.0338 *
T           -1.131e-01  2.852e-02  -3.968   0.0286 *
I(T^2)       5.430e-05  1.326e-05   4.097   0.0263 *
Multiple R-squared:  0.911,     Adjusted R-squared:  0.8517
F-statistic: 15.35 on 2 and 3 DF,  p-value: 0.02655
```

Figure 1. Output of R code. Equation, Coefficients, p-values, and Adjusted R-squared values are listed

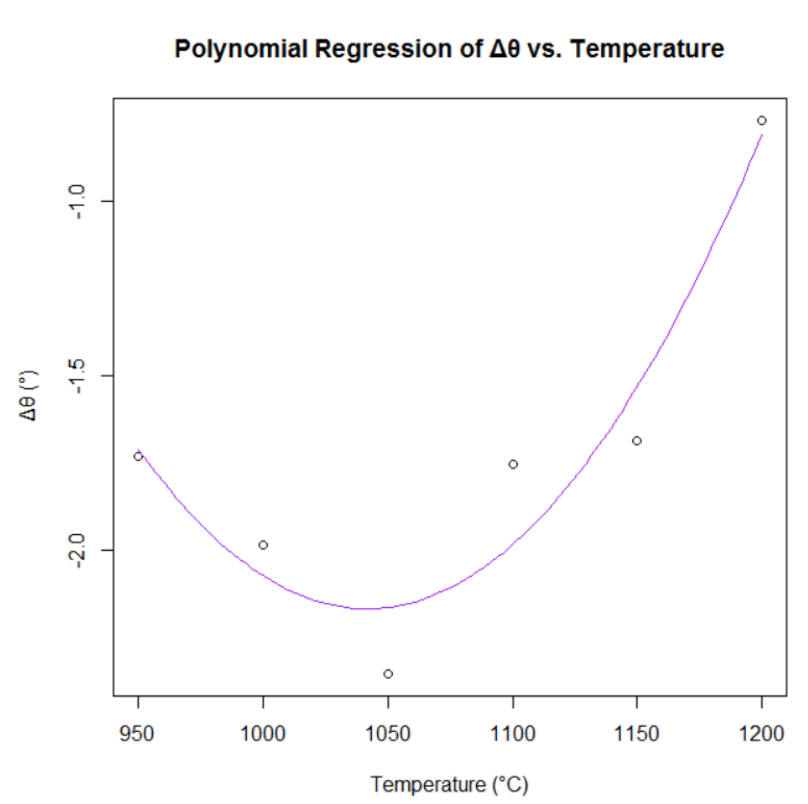

Figure 2. Plot of Δθ vs. Temp with generated curve of best fit. The curves minimum was used to determine the ideal fabrication firing temperature.

Methodology

Sensors were constructed from tape cast 8 mol% YSZ containing Au wire electrodes. They were fired for one hour at 1041°C, which was determined from the minimum of the Δθ vs. Temperature curve. A standard gas flow control system was used in conjunction with a Gamry Reference 600 in order to collect impedance measurements. Samples were tested at temperatures ranging from 600-700°C in an atmosphere of 1-18% O_2, 0-100ppm NO, and using N_2 to balance out the rest of the gases.

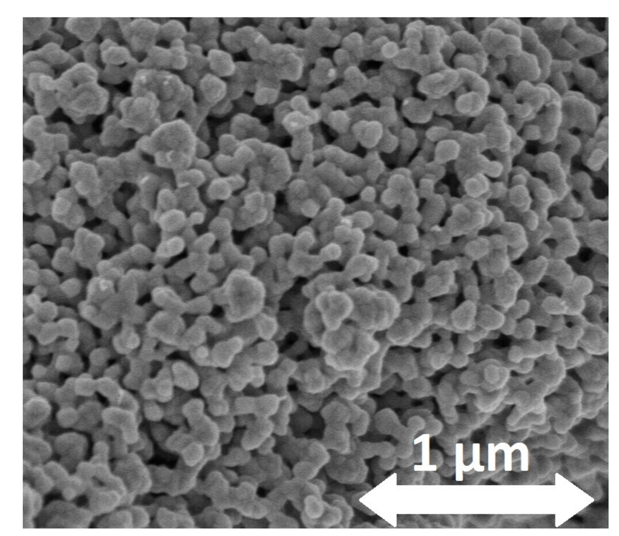

Figure 3. SEM image of YSZ electrolyte fired at 1041°C

Porosity calculations were made using two methods: Archimedes' method and utilizing Scanning Electron Microscope (SEM) images in conjunction with MATLAB computer software to approximate the porosity. For Archimedes' method samples had to be weighed while dry, saturated and submerged. Porosity was then calculated using the equation:

Porosity $= \Phi = (W_{sat} - W_{dry})/(W_{sat} - W_{sub})$

MATLAB analyzed the brightness of each pixel in the SEM image and then used the contrast of brightness between pixels to estimate the porosity of the YSZ electrolyte.[1]

Conclusions

Regression analysis using Δθ vs. Fabrication Temperature data was successful in identifying the ideal sensor fabrication temperature, 1041°C. Based on this analysis, sensors were fabricated. Analysis of fabricated sensors indicated a porosity of 54.8% which was determined via Archimedes' method. Computational methods via SEM images and MATLAB produced a porosity of approximately 46.76%. Further analysis is needed to confirm the porosity of the electrolyte more accurately. Analysis of the electrical response determined using impedance spectroscopy verified higher NO sensitivity was achieved.

Microstructural Results

Using the polynomial equation generated from R,

A TI-84 graphing calculator's 'minimum' function was used in order to obtain the minimum point on the curve. This point corresponds to the fabrication firing temperature of 1041°C.

Archimedes' method of porosity calculation yielded a value of 54.8%. The computational method via SEM images resulted in a porosity of approximately 46.76%. The large difference between each calculation can be associated to the methods themselves.

NO Sensitivity Results

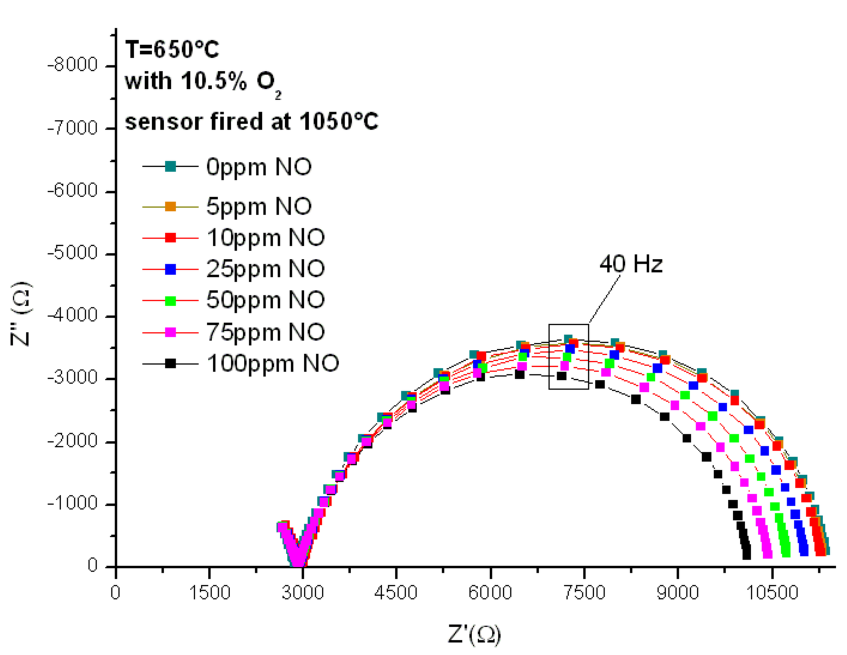

Figure 4. Typical Nyquist plot showing the electrical response of the sensor to NO.

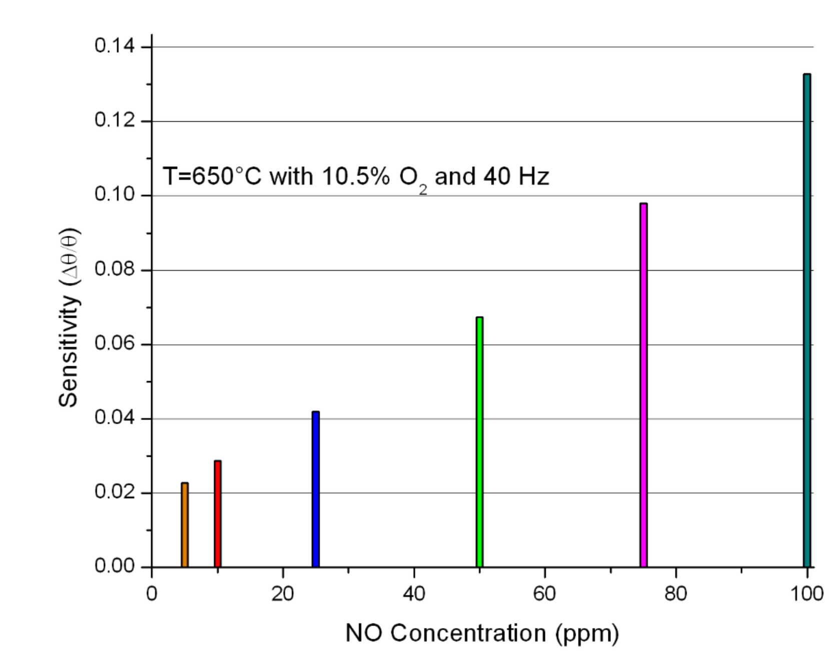

Figure 5. Plot of Sensitivity vs. NO Concentration.

Sensitivity data produced from sensors fired at 1041°C was slightly higher than sensitivity data from sensors fabricated at 1050°C in a prior study.[2] This confirms that the computational results identified the most suitable fabrication temperature for the NO_x sensor electrolyte microstructure.

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References

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2. L. Cui, F. Han, W. Dai and E. P. Murray, *Journal of Electrochemical Society*, **161** (3), B34-B38 (2014).