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# Thermopower and Conductivity of Aerosol Deposited Gas Sensitive $BaFe_{1-x}Ta_xO_{3-\delta}$ Films

*M. Bektas, T. Stöcker, G. Hagen, R. Moos*
*University of Bayreuth, Department of Functional Materials, Universitätsstraße 30, 95447 Bayreuth, Germany*
*E-mail: murat.bektas@uni-bayreuth.de*

**Abstract:**
In this study, the thermopower and the electrical conductivity of $BaFe_{1-x}Ta_xO_{3-\delta}$ sensor films were measured for *x* in the range of 0.1 to 0.45 (BFT*x*). Seebeck coefficient and conductivity were measured simultaneously between 400 and 850 °C under varying oxygen partial pressures from $10^{-2}$ to 1 bar. BFT*x* fine powders were prepared by a conventional mixed-oxide route. Crystal structure and phase purity were investigated by *X*-ray diffraction. BFT*x* thick-films were successfully deposited by the novel Aerosol Deposition Method (ADM) at room temperature on a special transducer. Both thermopower and electrical conductivity of samples with tantalum contents between $x = 0.2$ and 0.3 show almost no temperature dependency but dependent strongly on the oxygen partial pressure in the temperature range from 700 to 850 °C. Therefore making a combined, thermoelectric-conductometric sensing principle for temperature independent oxygen sensors is possible.

**Key words:** Aerosol deposition method, oxygen partial pressure, temperature independent oxygen sensor, Seebeck coefficient and conductivity, defect model.

## Introduction

Resistive high temperature oxygen sensors are discussed for many industrial applications [1]. The oxygen sensors should respond only to changes in the oxygen partial pressure but not to temperature variations [2]. $BaFe_{0.7}Ta_{0.3}O_{3-\delta}$ is known as a temperature independent conductometric oxygen sensor material with perovskite crystal structure [3]. The thermopower, also known as Seebeck coefficient, is an important parameter to determine constants for defect chemical models of semiconductor materials, since it is a measure for the charge carrier concentration [4]. The Seebeck coefficient does not depend on the geometry of the material [5], in contrast to the often investigated conductivity. The present work considers the thermopower as well as the electrical conductivity of $BaFe_{1-x}Ta_xO_{3-\delta}$ for *x* in the range of 0.1 to 0.45 (BFT*x*).

## Experimental Method

BFT*x* fine powders have been prepared by conventional mixed-oxide technique using $BaCO_3$ (99%, Alfa-Aesar), $Fe_2O_3$ (98%, Alfa-Aesar) and $Ta_2O_5$ (99.99%, Alfa Aesar), calcined at 1350°C in air. BFT*x* thick-films have been successfully deposited by the novel Aerosol Deposition Method (ADM) at room temperature on a special transducer in our department. Detailed information on ADM can be obtained from ref. [6]. The transducer includes four platinum electrodes and two gold-platinum thermocouples. For more information about the transducer the reader is referred to ref. [7]. The electrical conductivity was measured using a four point dc technique with a Keithley 2700 multimeter. Seebeck coefficient and conductivity measurements were performed simultaneously on the same sample between 400 and 850 °C under varying oxygen partial pressures from $10^2$ to 1 bar. One can determine the charge carrier densities and their mobility with the simultaneous conductivity and thermopower measurements [4].

## Results and Discussion

The electrical conductivity, $\sigma_h$, of a *p*-type (hole) conductor can be derived with Eq. (1). Here $\mu_h$ is the mobility of holes, *e* the elementary charge, and *p* the hole concentration.

$$\sigma_h = \mu_h \, e \, p \tag{1}$$

The thermoelectric power originating from holes $S_h$, for non-degenerate broad band semiconductor can be calculated using Eq. (2),

$$S_h = \frac{k}{e}\left(ln\frac{N_v}{p} + A_h\right) \tag{2}$$

where *k*, $N_v$ and $A_h$ refer to Boltzmann's constant, the effective density of states in the

valence band, and the transport constant for electron holes, respectively.

*X*-ray diffraction results show that neither BFT20 powder nor aerosol deposited thick-film have secondary phases. Since tantalum is a donor dopant, the conductivity of BFT*x* thick films decreases with increasing tantalum content. All samples have *p*-type conductivity in the investigated temperature and $pO_2$ range. Since, the samples with tantalum content between $x$ = 0.2 and 0.3 show almost temperature independent oxygen sensor behavior, we will discuss here the results of BFT20 aerosol deposited thick film sensor. Figure 1 shows the fast response of this sensor to the change of oxygen partial pressure. One can see the change of conductivity and $pO_2$ in the logarithmic scale with left and right *y*-axis, respectively. The sensor has temperature independent $pO_2$ dependency in the temperature range between 700 and 850 °C and exhibits a fast and reproducible response to $pO_2$ change even at a sensor temperature of 400 °C.

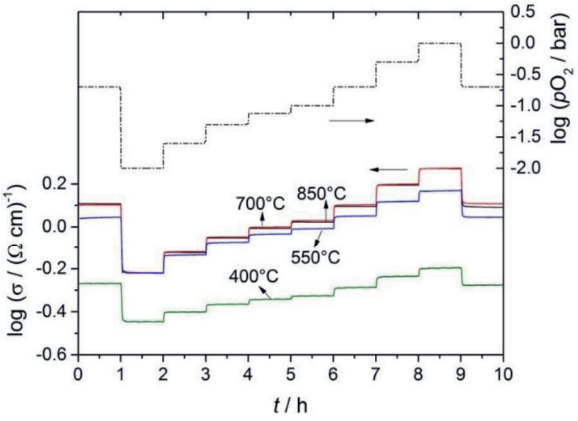


*Fig. 1. Logarithmic conductivity and $pO_2$ over time of a BFT20 aerosol deposited thick film. The curves for 700 °C and 850 °C are almost identical.*

Figure 2 and 3 show the oxygen partial pressure dependency of conductivity and thermopower of BFT20 sensor, respectively. Both, thermopower and electrical conductivity of the sensor show almost no temperature dependency but depend strongly on the oxygen partial pressure in the temperature range from 700 to 850 °C. The conductivity of the sensor follows oxygen partial pressure with a slope of 0.25. The oxygen sensitivity of the sensor decreases to 0.2 at 550 °C and the sensor is both, temperature and $pO_2$ dependent between 400 and 550 °C. One can see from Fig. 3 that the sample has positive Seebeck coefficient. No n-p-type transition was observed in the investigated oxygen partial pressure range. In contrast to the electrical conductivity, the Seebeck coefficient of all samples decreases with increasing oxygen partial pressure. All the other investigated BFT*x* ($x$ = 0.1 - 0.45) samples show the same behavior like BFT20. All of them have a positive Seebeck coefficients and their thermopower decreases with increasing $pO_2$.

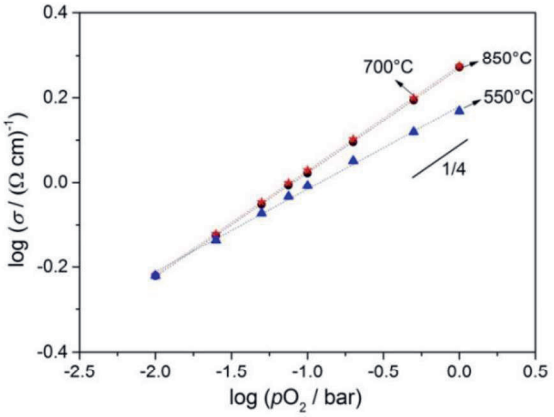


*Fig. 2. Log-log plot of the oxygen partial pressure vs. conductivity of a BFT20 aerosol deposited thick film. The curves for 700 °C and 850 °C are almost identical.*

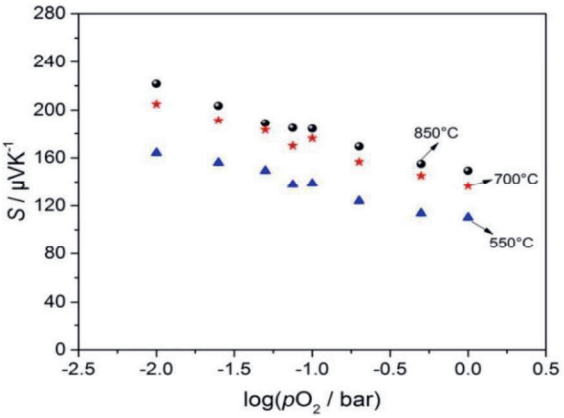


*Fig. 3. Oxygen partial pressure dependence of the Seebeck coefficient of a BFT20 aerosol deposited thick film.*

Using the standard defect chemical notation of Kröger-Vink [8], the defect chemical model of BFT20 can be written in high $pO_2$ range like in Eq. (3)

$$\sigma = (K'_{Ox}\,[A']/2)^{1/2}\; e\; \mu_0\; pO^{1/4} \exp^{(-E_a/kT)} \qquad (3)$$

Here $K_{Ox}$' is a temperature independent preexponential factor, [*A*'] the single ionized acceptor concentration, $\mu_0$ the preexponential factor of the hole mobility and $E_a$ the activation energy.

## Conclusion

The electrical conductivity and thermopower of BFT*x* ($x$ = 0.1 to 0.45) were simultaneously measured in different $pO_2$ ($10^{-2}$ to 1 bar) and at different temperatures (from 400 to 850 °C). The results show that aerosol deposited BFT*x* thick-films are *p*-type conductors in the investigated temperature and $pO_2$ range. Conductivity and thermopower of BFT*x* between $x$ = 0.2 and 0.3

depend strongly on the oxygen partial pressure change but are almost temperature-independent from 700 to 850 °C. Making a combined, thermoelectric-conductometric sensing principle for temperature independent oxygen sensors is possible, as for instance suggested in [5].

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