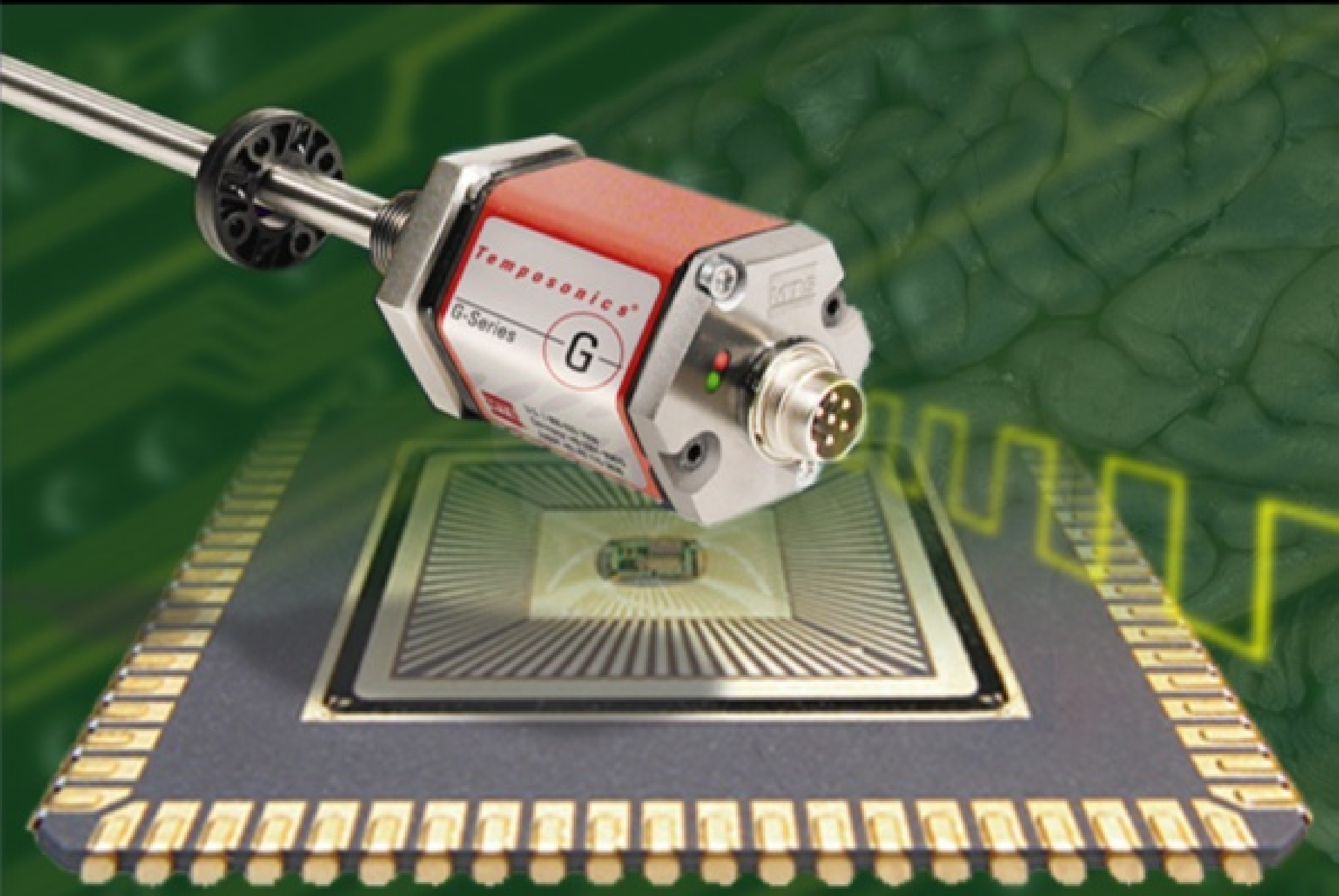
ISSN 1726-5749

SENSORS & TRANSDUCERS

vol. 77
3/07



Smart Sensors and Systems

International Frequency Sensor Association Publishing

IFSA

Sensors & Transducers

Volume 77
Issue 3
March 2007

www.sensorsportal.com

ISSN 1726-5479

Sensors & Transducers Journal (ISSN 1726-5479) is a peer review international journal published monthly online by International Frequency Sensor Association (IFSA). Available in electronic and CD-ROM.

Sensors & Transducers Journal

Contents

Volume 77
Issue 3
March 2007

www.sensorsportal.com

ISSN 1726-5479

Research Articles

Authors are encouraged to submit article in MS Word (doc) and Acrobat (pdf) formats by e-mail: editor@sensorsportal.com
Please visit journal's webpage with preparation instructions: http://www.sensorsportal.com/HTML/DIGEST/Submition.htm

Sensors & Transducers Journal, Vol.77, Issue 3, March 2007, pp.1025-1031

Sensors & Transducers

ISSN 1726-5479

http://www.sensorsportal.com

High Temperature Humidity Sensor for Detection of Leak Through Slits and Cracks in Pressurized Nuclear Power Reactor Pipes

Debdulal Saha, Kamalendu Sengupta*

Sensors and Actuators Section, Central Glass & Ceramic Research Institute, 196 Raja S. C. Mullick Road, Kolkata-700032, India

Tel.: 091-33-2473-3496/69/76, fax: 2473-0957

*E-mail: kamalendusengupta@yahoo.co.in

Debdulal76@yahoo.co.in

Received: 10 February 2007 / Accepted: 15 March / Published: 26 March 2007

Abstract: The leak before break (LBB) concept is well known to nuclear power reactor. The problem is common to water power reactor. This is based on the premise that a detectable leak will develop before catastrophic break occurs. The main purpose of the present study is to develop tape cast $MgCr_2O_4$+35mole% TiO_2 and gel cast γ-Al_2O_3 humidity sensor for use in LBB applications at 300^0C. The material capacitance varies with transient injection of water vapour adsorption. In actual plant, the sensors are placed on a steam pipe surrounded by heat insulation. The pipe unites the nuclear reactor and power generator. The analysis of humidity distribution in the annulus is calculated assuring leak rate 0.1gpm in a 30 m long tube. In this paper, analysis is done on the basis of the two types of sensor using AC frequency. Performance characteristics are observed for the LLB application.

Keywords: High temperature moisture sensor, Tape cast, Nuclear power reactor

1. Introduction

The choice of Mg_2CrO_4-TiO_2 tape cast as sensor elements in the form of porous thick films is considered. The inherent hydrophilicity to water molecules, Mg_2CrO_4-TiO_2 based humidity sensors responses of surface conductance under an electric field, when the environment humidity is low; the protons released from surface hydroxyls help from site to site from the spinel surface or hydrate with adjacent water molecules to form H_3O^+ ions as charge carriers. At high humilities the vapour molecules inside the pores form a continuous layer, which enhance transfer of protons. The sensing response depends on pore geometry of spinel structure. Mg_2CrO_4-TiO_2 spinel structure is effective for H_3O^+ ions as charge carriers [2-3, 6]. The study presents new results for electrical sensing properties of Mg_2CrO_4-TiO_2 spinel and aero gel alumina thick films after depositing on parallel gold electrodes. To detect leakage, humidity sensors are used for sensing of moisture through slits and cracks in pipes for nuclear power reactor. If steam leak occurs in reactor pipe, there is a possibility of reactor failure and the radiation leaks into the containment area environment. The sensors are required to withstand significant amounts of heat at 300°C [1-3] without failure in the event a leak occurs into the containment area. Such a humidity sensor is already implemented in European Nuclear Power Plants [4-5]. According to NUREG-1061 (U.S.N.R.C, 1984) [1] the essentials of a leak sensor are as follows:

1) Leak detection system should be able to detect 1gram per minute (gpm) in the identified leak, and 0.1 gpm in the unidentified leak

2) The leak detection systems must have three diversity, each with three redundancies.

3) Local sensors are applicable.

Here in this paper the electrical characteristics of humidity sensors have been described for use directly in Main Stream Line (MSL) piping.

2. Experimental

Stoichiometric amount of nitrate salts were dissolved separately in distilled water, then a stoichiometric amount of citric acid was added and after the complete mixing a homogenous solution was achieved. These solutions were slowly evaporated at around 80-100°C. In the system dehydration takes place followed by gelation to form highly viscous colloidal mass but the gelation probably results due to the formation of Metal-citrate-nitrate complex. On continuous heating the mass was converted in to ash. This ash was calcined at 900°C for 4 hours. The end product of the synthesis was $MgCr_2O_4$. The crystalline phase was identified at room temperature by standard x-ray diffraction technique using Cu- radiation with a Philips PW 1730 diffractometer. Humidity Sensor ($MgCr_2O_4$+35mole%TiO_2) thick tape cast is sensitive to humidity at 300°C [6] and fit for MSL operational condition. The change in AC conductivity can be measured by a low cost electronic device. Both $MgCr_2O_4$+35mole%TiO_2 and γ-Al_2O_3 are sensitive to H_2O ionic condition [7-8]. Preparation of characterization of material γ-Al_2O_3 was prepared by Yoldas method [11].

3. Results and Discussion

SEM (SEM LEO, S43Oi, LEO, UK) micrograph shows surface structure contains high degree of porosity with a large enhancement of active surface, responsible for the high absorption and adsorption ability of the material. γ-Al_2O_3 phase crystallographic peak and $MgCr_2O_4$-35mole%TiO_2 crystallographic peak were done by XRD PW 1730 using Cu Kα radiation (Philips, Holland).

3.1 SEM Picture:

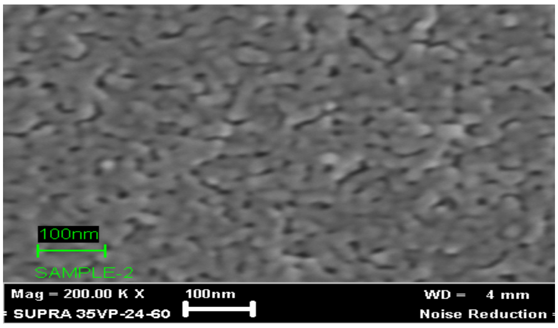


Fig. 1. Nanoporous γ-Al_2O_3 tape.

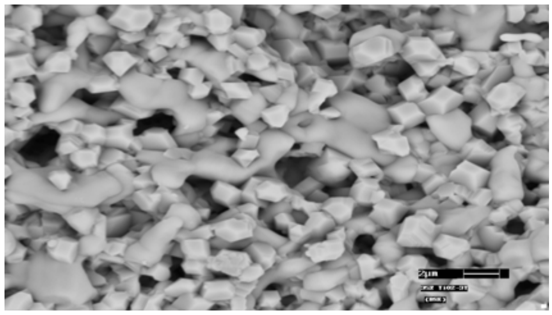


Fig.2. Micro porous tape cast $MgCr_2O_4$+35mole% TiO_2

3.2 Sensor Picture.

Fig. 3 shows the complete structure of a nano porous sensor, electrode on both sides. Fig. 4 shows micro porous structure with parallel electrodes fixed on alumina strips. The strips are provided with conducting tracks for inserting it into a temperature zone of 300^0C. The 75 percent of the strip will remain in the annalus portion and the rest 25 percent will remain out side the insulation for connection to active device.

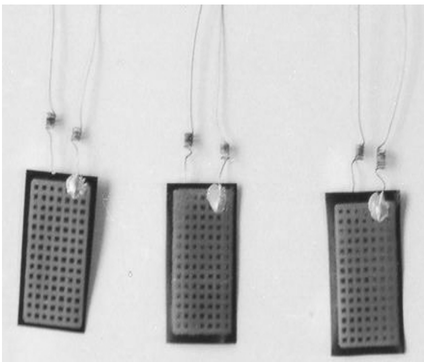


Fig. 3. γ- Al_2O_3 sensor.

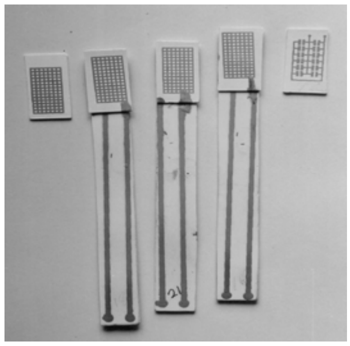


Fig. 4. $MgCr_2O_4$ +35mole%TiO_2 sensor.

3.3 XRD-analysis.

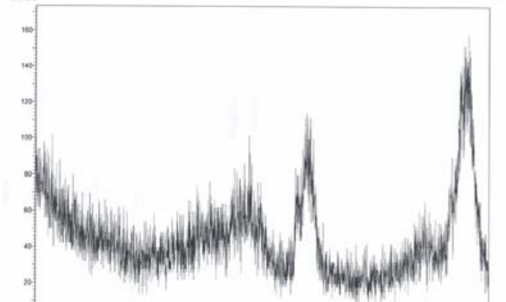


Fig. 5. γ- Al_2O_3

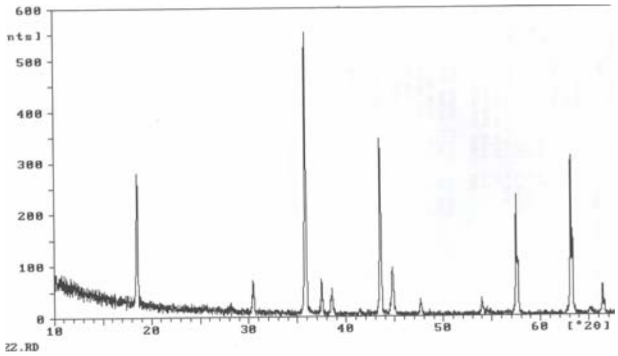


Fig. 6. $MgCr_2O_4$ –35 mole%TiO_2

3.4 Impedance Gain Phase Analysis Data for MgCr2O4 +35mole%TiO2 & γ -Al2O3:

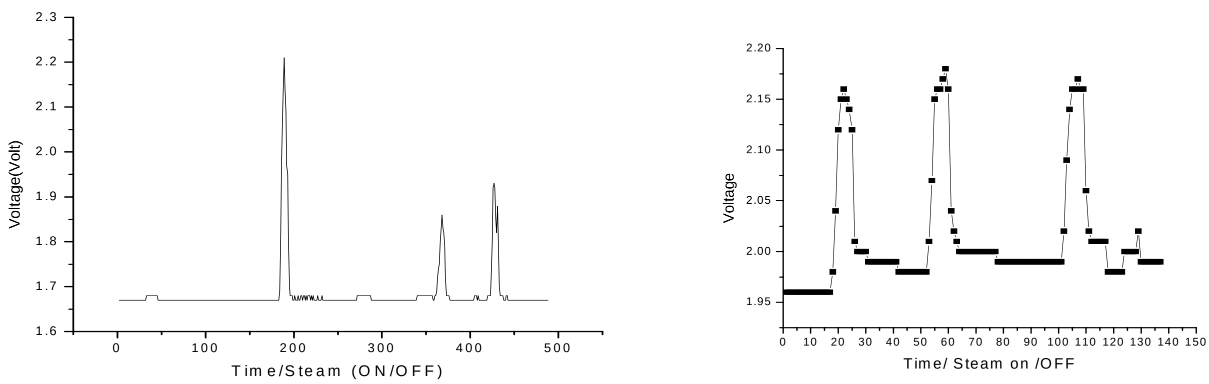
Table 1.

Fr(Hz)	RH(%)	Cp	D	X (Ω)	Z (Ω)	θ (theta) δεγ.
1000	0	100pF	0.11	-1.5M	1.5M	-83
1000	85	756pF	2	-61.3k	79.5k	-39.9
1000	0	90pF	0.054	-1.4M	1.8M	-86.94
1000	85	86nF	>10	-36k	828k	-2.5

The conduction mechanism of $MgCr_2O_4$+35mole%TiO_2 tape cast start in a way such that the absorbed fist layer of water molecules dissociate hydroxyl ion (OH^-) and proton (H^+), H^+ chemisorbed on the surface metal ion combines with surface oxygen to form second OH^- ions. Chromium is most active surface metal ion because Cr^{3+} easily combines with OH^- ion. The weakly bonded hydroxyl group dissociates to form mobile protons. Mobile protons on the surface can migrate by hopping to increase the electrical conductivity (Anderson and Parks[5] 1998). It is seen from the table 1 that when water is injected to increase the humidity level, Z changes from 1.5 MΩ to 79.2 KΩ**.** The response of the tape material with time in MSL operation at 300º C is shown in Fig. 7a. Dissipation factor increases due to proton hopping and ionic conduction [4]. That from equation,

$$\in^* = \in - j\in'' = \in - j\,(k/\omega\in_o) \qquad (1)$$

The overall capacity changes to a high value are shown in Table 1. Intermittent rise of capacitance Fig. 7a is due to intermittent insertion of steam in the insulation sheath. In case of Al_2O_3 shown in Table 1 the change in Z value from 1.8 M to 828 K from low to high humidity condition and dissipation factor changes from 0.054 to >10, Cp changes from 90pF -86nF. So γ-Al_2O_3 can be considered as a good candidate for detection of leakage in water Reactor. For long time operation γ-Al_2O_3 may be considered as an ideal sensor for its highly crystallographic stable structure. The change in sensor voltage with steam injection is shown in Fig. 7 b.



(a) (b)

Fig. 7. Transient response of (a) $MgCr_2O_4$+35mole%TiO_2 and (b) γ-Al_2O_3 during steam injection measured at temperature T=300^{0}C.

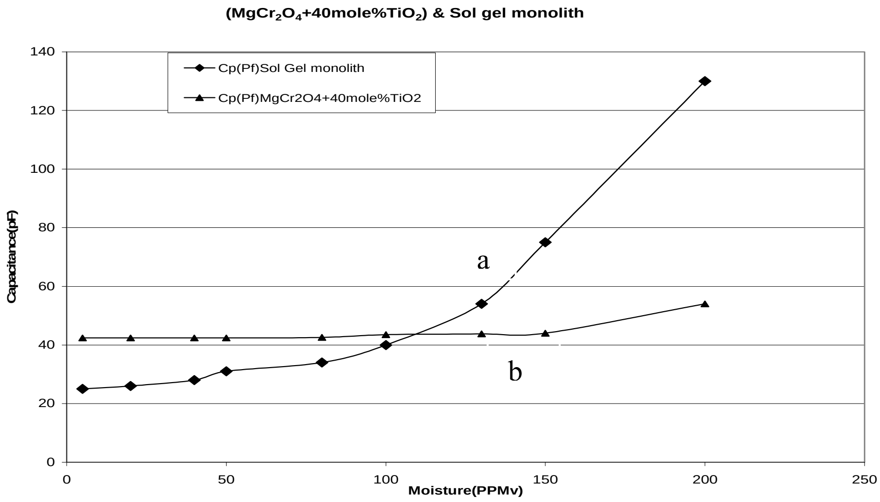
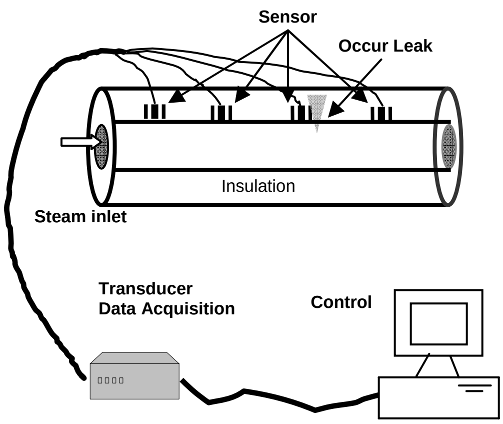


Fig. 8. Study of sensitivity (a) γ-Al_2O_3 and (b) $MgCr_2O_4$+35 mole%TiO_2 in trace moisture level.

From the theoretical analysis the generation of moisture is calculated per second. It is clear that nano porous γ-Al_2O_3 is sensitive to trace moisture than $MgCr_2O_4$+35 mole%TiO_2.

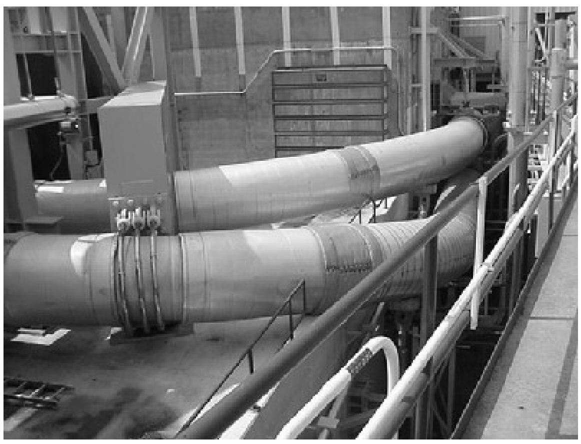
4. Location Analysis

Leak is postulated to occur as a line source representing a crack. The concentration of moisture at a distance X from the crack line at a time (T) can be determined by the experimental arrangement as shown in the Fig 9(a). A 30 m length tube of 30-inch diameter is covered by 8 mm insulation sheath of wood fiber. 20 sensors were installed at equal spacing with a separation of 1.5 m. Each sensor was concentrated to a 20-channel Data Acquisition system.



a)

Fig. 9. Schematic diagram of location analysis.



b)

Fig. 9. Nuclear Reactor pipe.

Table 2.

Channel/sensor position	**Successive Distance from left side of the pipe (m)**	**Voltage (mV)**	**Time(T)**
1	1.5	8	< 1 min
2	3	12	-do-
3	4.5	14	-do-
4	6	15	-do-
5	7.5	16	-do-
6	9	17	-do-
7	10.5	18	-do-
8	12	20	-do-
9	**13.5**	**28**	**-do-**
10	**15**	**30**	**-do-**
11	**16.5**	**27**	**-do-**
12	18	20	-do-
13	19.5	15	-do-
14	21	14	-do-
15	22.5	10	-do-
16	24	8	-do-
17	25.5	7	-do-
18	27	6	-do-
19	28.5	3	-do-
20	30	2	-do-

This is a straightforward method of location analysis. From Table 2 the location of steam leakage lies between 13.5 m to 16.5 m from channel 1, first sensor from left of the pipe in the array of 20 sensors. Response time of the sensors is within one minute so the position of maximum voltage/min. is the probable leakage position.

5. Conclusion

$MgCr_2O_4$+35mole% TiO_2 andγ-Al_2O_3 sol-gel template/tape has been developed to detect pipe cracks/slits in a pressurized system. The sol-gel based nanoporous γ-Al_2O_3 template has adequate sensitivity over the leak rate 200 ppm per minute over the temperature 300^{o}C. AC response is found to be useful for humidity detection. For LBB realization parallel connection method with AC voltage measurement is proposed. By parallel connection system both leak rate and leak position can be detected accurately. The result shows that ceramic humidity sensor can be used for leak detection and position of leak for use in Nuclear power plant.

Acknowledgement

The authors are thankful to Board of Research in Nuclear Sciences, Department of Atomic Energy, Govt of India for financial support and the authors also thankful to Dr. H. S. Maiti, Director, Central Glass and Ceramic Research Institute, Kolkata for rendering all sorts of cooperation for conducting the research work. The authors are sincerely appreciated the kind help and assistance of Mr. A.K.Mandal, Dr. Mrs S. Sen in doing the SEM experiments at CGCRI.

References

[1]. US. NRC, 1984, *Evaluation of potential for pipe Breaks*, Nureg, 1061, Vol. 3.
[2]. Na Young Lee, II Soon Hwang, Chang Rock Song and Han II Yoo, Development of ceramic humidity sensor for the application of the Leak-before-Break concept, *Transaction of the 15th International Conference on Structural Mechanics in Reactor Technology (SMIRT-50)*, Seoul, Korea, August 1999, pp. 15-20.
[3]. Na Young Lee, II Soon Hwang, Chang Rock Song and Han II Yoo, New Leak detection Technique using ceramic humidity sensor for water reactors, *Nuclear Engineering and Design,* 205, 2001, pp. 23-33.
[4]. U.S. NRC, Reactor coolant Pressure Boundary Leakage detection systems, Regulatory Guide 1 45, Rev. 1. 1982.
[5]. P. Jax, Detecting and locating the smallest leaks early, *Nuclear Engineering International*, 1995, pp. 22-24.
[6]. T. Nitta, Z. Terada and S. Hayakawa, Humidity-Sensitive Electrical Conduction of $MgCr_2O_4$–TiO_2 Porous Ceramics, *J Am Ceram. Soc.,* Vol. 63, No [5-6], 1980, pp. 295-300.
[7]. K. Mistry, D. Saha, K. Sengupta, Sol-Gel processed alumina thick film template as sensitive, capacitive trace moisture sensor, *Sensors and Actuators B* (accepted Aug. 04), 2004.
[8]. Anderson, J.H. Parks, GA, The electrical conductivity of silica gel in the presence of absorbed water, *J. Physics Chem* 72 (10), 1968, p. 3662-3668.
[9]. Z. M. Rittersma, Recent achievements in miniaturized humidity sensors - a review of transduction techniques, *Sensors and Actuators A* 96, 2002, pp. 196-210.
[10]. Pieter R. Wiederhold, Water vapor measurement-Methods and Instrumentation, Marcel Dekker Inc., New York, Table 13.7, 1997, pp 299.
[11]. B.E. Yoldas, A transparent porous alumina, *Jr. of Mater. Sci.*, 10, 1856 (1975).

___

Guide for Contributors

Aims and Scope

Sensors & Transducers Journal (ISSN 1726- 5479) provides an advanced forum for the science and technology of physical, chemical sensors and biosensors. It publishes state-of-the-art reviews, regular research and application specific papers, short notes, letters to Editor and sensors related books reviews as well as academic, practical and commercial information of interest to its readership. Because it is an open access, peer review international journal, papers rapidly published in *Sensors & Transducers Journal* will receive a very high publicity. The journal is published monthly as twelve issues per annual by International Frequency Association (IFSA). In additional, some special sponsored and conference issues published annually.

Topics Covered

Contributions are invited on all aspects of research, development and application of the science and technology of sensors, transducers and sensor instrumentations. Topics include, but are not restricted to:

- Physical, chemical and biosensors;
- Digital, frequency, period, duty-cycle, time interval, PWM, pulse number output sensors and transducers;
- Theory, principles, effects, design, standardization and modeling;
- Smart sensors and systems;
- Sensor instrumentation;
- Virtual instruments;
- Sensors interfaces, buses and networks;
- Signal processing;
- Frequency (period, duty-cycle)-to-digital converters, ADC;
- Technologies and materials;
- Nanosensors;
- Microsystems;
- Applications.

Submission of papers

Articles should be written in English. Authors are invited to submit by e-mail editor@sensorsportal.com 4-12 pages article (including abstract, illustrations (color or grayscale), photos and references) in both: MS Word (doc) and Acrobat (pdf) formats. Detailed preparation instructions, paper example and template of manuscript are available from the journal's webpage: http://www.sensorsportal.com/HTML/DIGEST/Submition.htm Authors must follow the instructions strictly when submitting their manuscripts.

Advertising Information

Advertising orders and enquires may be sent to sales@sensorsportal.com Please download also our media kit: http://www.sensorsportal.com/DOWNLOADS/Media_Kit_2007.PDF