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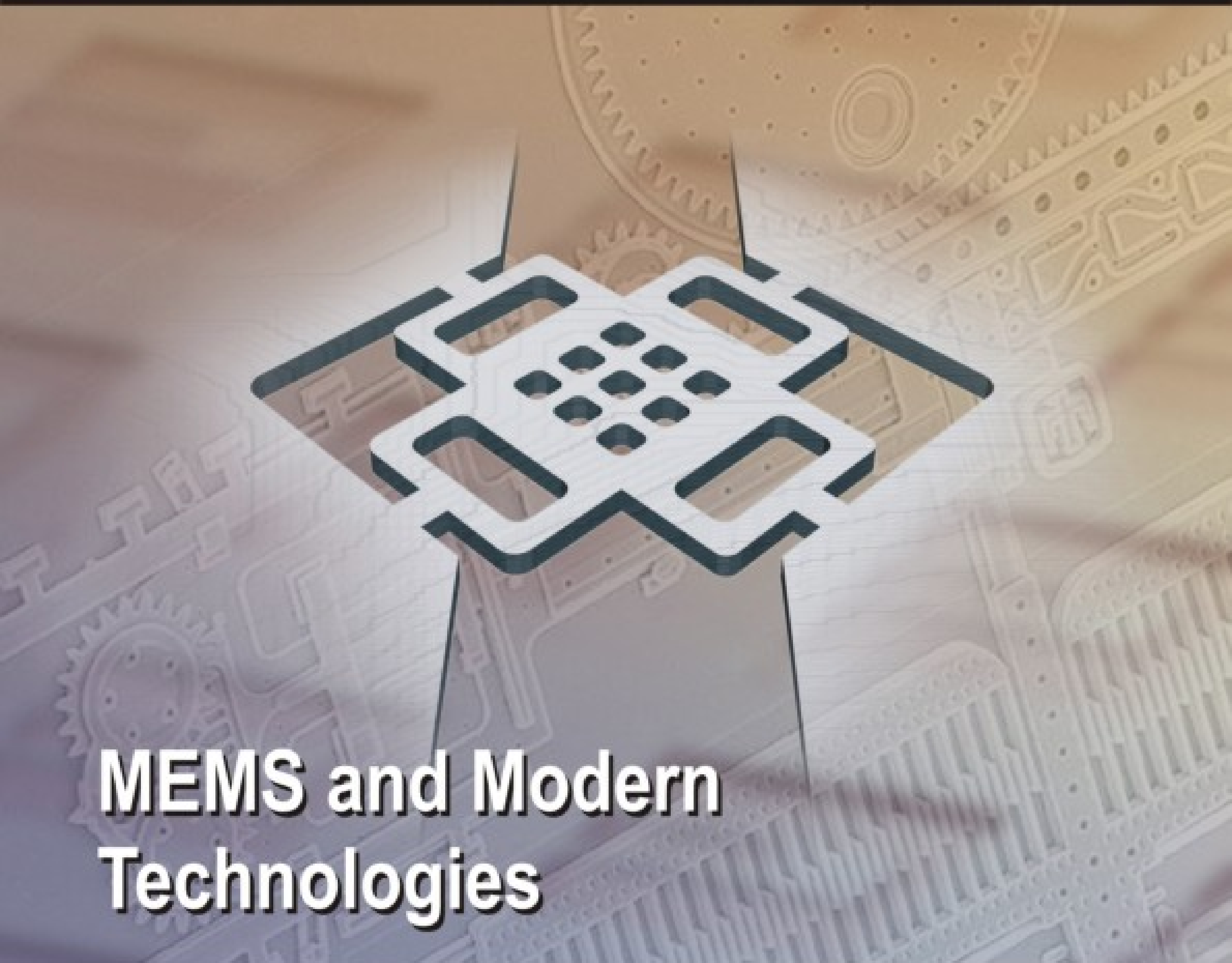

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Fabrication of Meso-Porous Gamma-Alumina Films by Sol-Gel and Gel Casting Processes for Making Moisture Sensors

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Abstract: Meso-porous γ-Al_2O_3 film may be used as a highly sensitive trace moisture sensor. The crack-free alumina film was developed using a combination of sol-gel and tape casting processes, which produce high porosity, high surface area and small pore dimensions in the range of few nano-meter at uniform distribution. Sol-gel processes are well known in nano-technology and nano-material preparation, but it is difficult to make crack-free thick or thin films using this method. Tape cast methods are used for the fabrication of flexible crack-free thick ceramic sheets. Our objective was to develop nano-structured, crack-free, transparent Al_2O_3 film a few microns thick, has a highly porous and stable crystallographic nature. A metallic paste was printed by screen printing on both side of the film surface for electrodes to form a sensitive element. A silver wire (dia φ=0.1mm) lead was connected to a grid structure electrode using a silver paste spot for fine joining. Alumina is absorbs moisture molecules into its meso-porous layer and changes its electrical characteristics according to the moisture content, its dielectric constant increase as moisture increase. Moisture molecules can be conceived of as dipoles in random state before the application of an electric field. When the dipole orientation was changed from random to an equilibrium state under the application of external field, a large change in dielectric constant was observed. The number of water molecules absorbed determines the electrical impedance of the capacitor, which in turn is proportional to water vapor pressure. *Copyright © 2007 IFSA.*

Keywords: Gel casting, γ-Al_2O_3 film, Moisture sensor.

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1. Introduction

Sol-gel technology has attracted increasing attention in recent years, for scientific and technological applications because of its low cost, the case of fabricating of films or coatings of complex oxides, and the ability to control composition and microstructure of nano-materials. On the other hand, the sol-gel process is suitable for preparing porous ceramics[1] because the reaction proceeds with a co-existent solvent phase, which the evaporation leaves cavities[2] in the materials. One special interest has been preparing homogeneous, transparent flexible sheets without macroscopic cracks[3] formed during drying and heating stages. Different organic compounds such as binders, plasticizers, homogenizers were mixed stepwise into a gel to make slurry which is then cast to make flexible[3], uniformly thick, crack-free transparent[3-5] sheets. Sheets may be cut in to any shape and size according to the design of the electrode on the film surface, which may be done very easily at the green stage by screen printing. Finally, the film was cured slowly increasing the temperature and soaking to remove burnt organic compound, to form alumina with the crystallographic face and cure the electrode. As a result, sensor preparation time and expenses are reduced.

2. Fabrication of Film

Boehmite gel was prepared using Al-sec-butoxide ($C_{12}H_{27}AlO_3$) as a staring material[5-7] and reaction was carried outdone by adding excess water at 90-100^oC the butoxide to water molar ratio was 1:100) with vigorous stirring for 1-hour to produce a colloidal solution of aluminum hydroxide after reaction and evaporation of butanol ($C_4H_{10}O$). Hydroxide molecules were ionized by peptization by adding hydrochloric acid (HCl) acid until the colloidal became transparent. Generally $Al(OH)_3$ is a positive ion acceptor, and in the presence of HCl acid each hydroxide accepts an H^+ ion and becomes $Al(OH)_3^+$. Because the charges, the ions repeal each other and disperse at the molecular level and as a result particle is reduced to a very smaller size of about a few nanometers. For completing the peptization takes another 1-hour at 100^oC to form a transparent stable solution. The solution was then refluxed for about 16-hours in a refluxing bath at 100-110^oC, and a stable boehimite sol of nano-particles was prepared. After evaporating excess water from the sol, sol was dried in to the air dryer to increase the suitable density to approximately 1.01-1.04gm/cc, and the viscous gel was ready for tape casting process.

Polyvinyl alcohol (PVA), of molecular weight 125,000, binder was added to the prepared gel at a weight ratio of 1gm gel with 16.7 mg PVA and stirred for 2-hours at 60^oC to insure uniform mixing of the PVA into the gel. It was then cooled to room temperature, and benzylbutylphthalate (BBP), chemical formula is $C_{19}H_{20}O_4$, molecular weight 31,237gm and polyethylene glycol (PEG), molar weight 380-420gm, added in to the slurry at a weight ratio of 1gm sol with 0.5mg BBP and 0.007mg PEG, as plasticizer and homogenizer respectively. Mixed slurry was stirring at room temperature 25^oC for 4-hour in a vacuum chamber. The slurry was bubble-free and was used to making the tape. The slurry was cast on a flat glass plate with a blade gap of 2.5 mm adjusted so that after air-drying the thickness of the green tape was 110-120 μm. It was then removed the plate as a flexible thick sheet (Fig. 1). The sheet was tested by XRD**(** PW 1730, Philips) shown in Fig.2, chemical change and crystal formation. The 1st major peak denoted where $Al(OH)_3$ transform to pure α-Al_2O_3 at about 320^oC temperature and next peak denoted phase transformation from α phase to γ-phase temperature at about 484^oC**.**

Fig.1. Transparent flexible boehmite sheet.

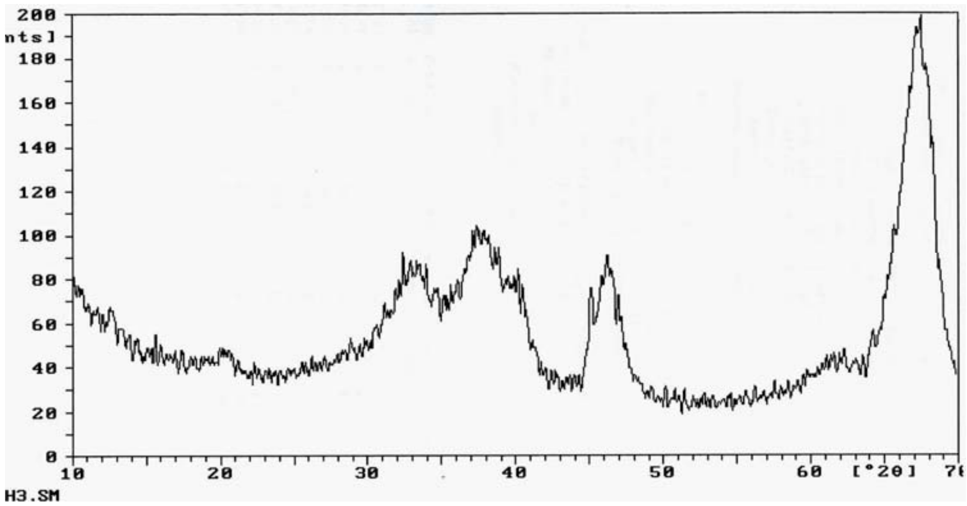

Fig. 2. XRD for determination of γ-phase alumina peak fired at temperature to 900°C.

3. Fabrication of Electrode

AgPd conductive paste was screen printed on fabricated flexible sheets with suitable designs and cut according to the electrode surface area. Electrode film fired using the same stepwise temperature program up to 850°C-900°C, and parallel plate capacitor type sensor[14-18] was formed (Fig. 3) for testing and electrical characterization. Surface structure and thickness of the sensor were determined using SEM (LEO, S43Oi, LEO, UK) micrographs shown in Fig. 4 and Fig.5.

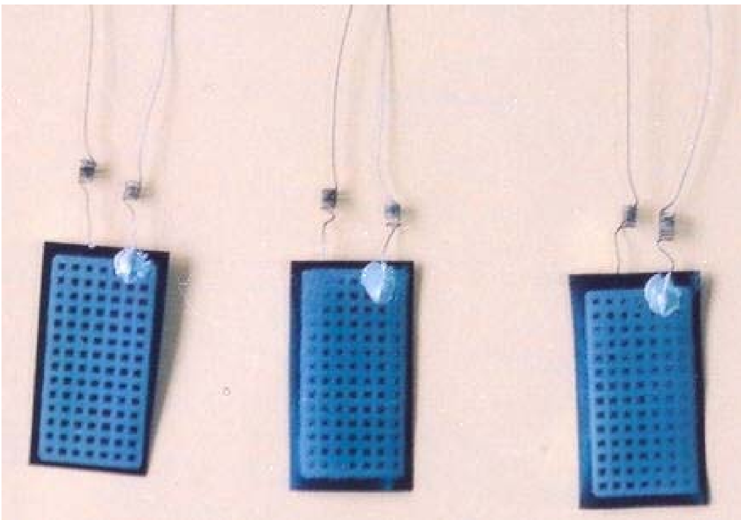

Fig. 3. Complete sensor with lead wire.

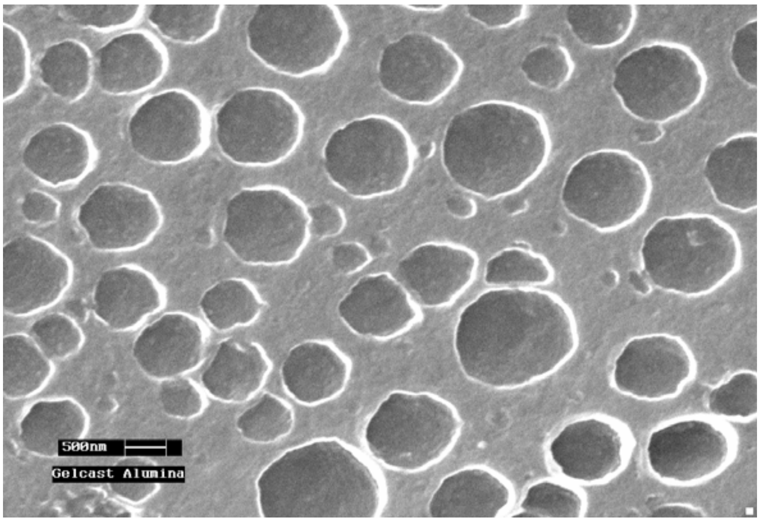

Fig.4. SEM image showing for surface pore structure.

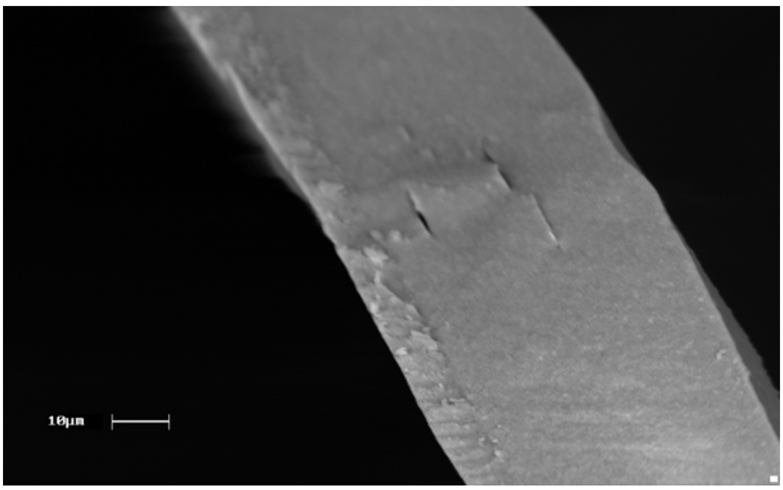

Fig. 5. SEM image to determine film thickness.

4. Experimental Set-Up

The electrical performance of the sensor was examined by placing it in a stainless steel chamber with a volume of 250 cm^3 and held at 25^o C within a thermostatic housing. The desired ppm was obtained by mixing dry nitrogen gas with the proper amount of moist nitrogen gas using a needle valve to control the in known concentration of the moisture. The mixed gas flow rate was set to 3-4 ltr. min^{-1}, gas allowed to flow into the test chamber, and the presence of moisture was monitored by a standard commercial sensor from SHAW(SADPTR-R).

Sensor leads were connected to an HP precession LCR meter (HP4284a) to measure sensor capacitance, resistance, impedance, loss factor, phase angle at different moisture concentrations valued observed as shown in Table 1.

Table 1.

Moisture (ppm)	**Capacitance (pF)**	**Loss**	**Impedance (MΩ)**	**Phase (Deg)**
10	47.7	.035	3338	88.03
30	59.3	.204	2629	78.44
50	72.3	.497	1973	63.5
56	79.6	.684	1647	55.51
70	110.5	1.595	765	32.09
90	168.6	3.687	246	15.14

Characteristics curves in (Fig. 6, 7, 8 and 9) show significant change of capacitance, impedance, loss factor and phase shift occurring due to very small changes in moisture concentration at a frequency 1-KHz. It was also observed that the response and recovery times are shorter by applying sudden changes in moisture content from 0-200ppm and reverse. The thickness of the films is optimal for handling and tolerating all steps in the preparation of the device (for example, the electrode deposition and presents good mechanical properties. A great variety of γ-Al_2O_3 film have been employed in the search for application as sensor[12-18] of moisture at the ppm level. The potentialities of γ-Al_2O_3 gel cast films as the sensitive element in a dew point meter was evaluated here for the first time. The ease of synthesis, capabilities of the film, and possibility of application are a good starting point, together with the high absorption ability and chemical inertness typical of this kind of sol-gel monolith. The great porosity shown by the Al_2O_3 monolith gives rise to an extended surface that is active in the absorption of water vapor.

As a consequence of the variation in capacitance and resistance calculate an equivalent circuit of sensor by modeling the electrical behavior. At a constant frequency it can be suitable for a complete device application for industrial need and also have a commercial value.

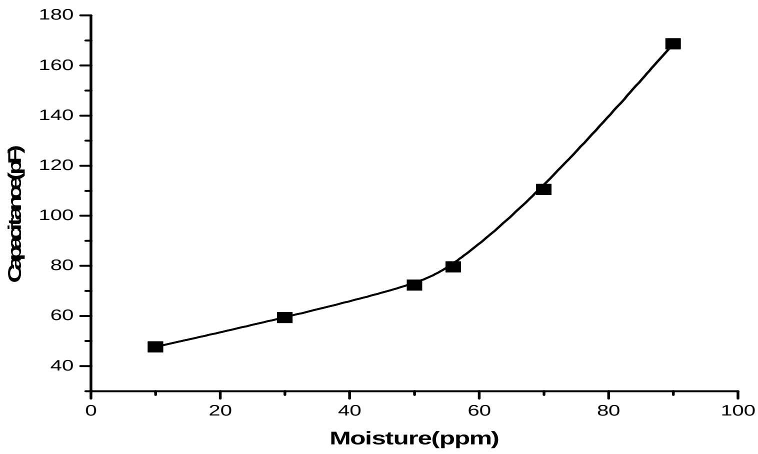

Fig. 6. Capacitance versus moisture plot at 1 KHz.

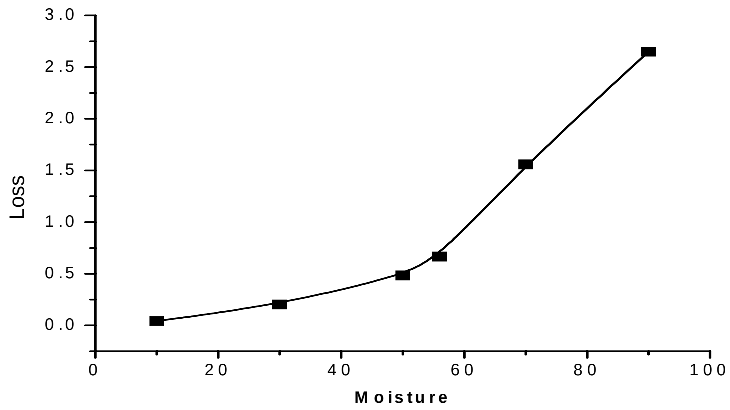

Fig. 7. Loss versus moisture plot at 1 KHz.

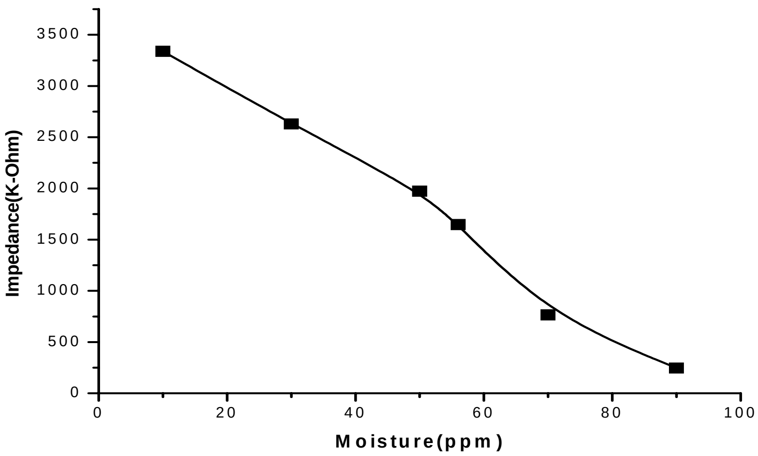

Fig. 8. Impedance versus moisture plot at 1 KHz.

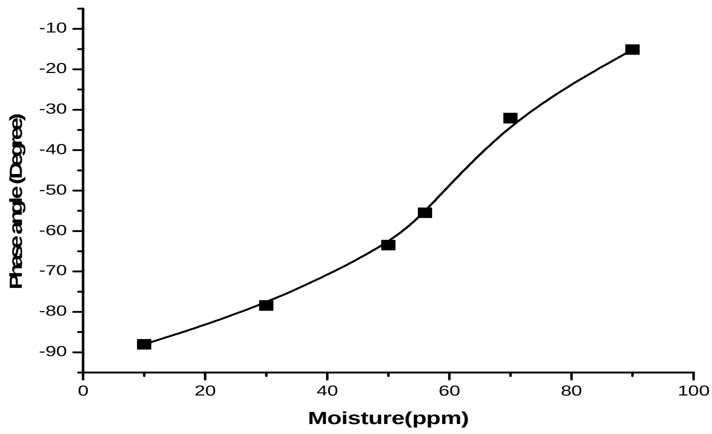

Fig.9. Phase shift versus moisture plot at 1 KHz.

5. Conclusions

Porous γ-Al_2O_3 film has been fabricated using a standard sol-gel process combined with tape casting. An electrode may be printed on the film surface at the green stage easily with any shape and design due to the flexibility of the sheet. The sensor acted as the sensing dielectric for trace amounts of moisture due to its highly porous surface. The sensitivity of the sensor was 1 pF per 4 ppm. The reaction proceeds with co-existence solvent phases, the evaporation of which evaporation leaves cavities in the material.

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Guide for Contributors

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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.

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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:

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- Microsystems;
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