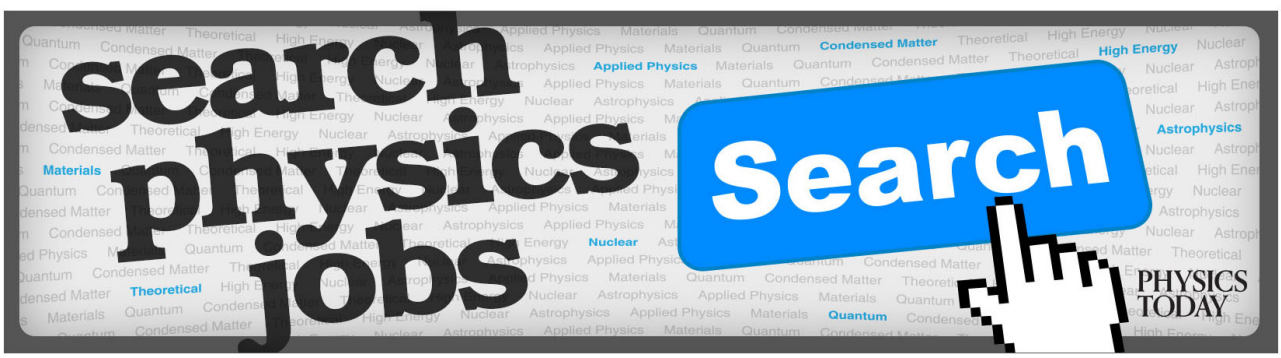
AIP | Review of Scientific Instruments

Note: Real time optical sensing of alpha-radiation emitting radioactive aerosols based on solid state nuclear track detector

A. Kulkarni, S. Ha, P. Joshirao, V. Manchanda, M. S. Bak, and T. Kim

Citation: Review of Scientific Instruments **86**, 066101 (2015); doi: 10.1063/1.4922463
View online: http://dx.doi.org/10.1063/1.4922463
View Table of Contents: http://scitation.aip.org/content/aip/journal/rsi/86/6?ver=pdfcov
Published by the AIP Publishing



Note: Real time optical sensing of alpha-radiation emitting radioactive aerosols based on solid state nuclear track detector

A. Kulkarni,[1,a)] S. Ha,[2,a)] P. Joshirao,[3] V. Manchanda,[3] M. S. Bak,[1,b)] and T. Kim[1,2,b)]
[1]*School of Mechanical Engineering, Sungkyunkwan University, Suwon 440-746, South Korea*
[2]*SKKU Advanced Institute of Nano Technology (SAINT), Sungkyunkwan University, Suwon 440-746, South Korea*
[3]*Department of Energy Science, Sungkyunkwan University, Suwon 440-746, South Korea*

(Received 9 March 2015; accepted 1 June 2015; published online 11 June 2015)

A sensitive radioactive aerosols sensor has been designed and developed. Its design guidance is based on the need for a low operational cost and reliable measurements to provide daily aerosol monitoring. The exposure of diethylene-glycol bis (allylcarbonate) to radiation causes modification of its physico-chemical properties like surface roughness and reflectance. In the present study, optical sensor based on the reflectance measurement has been developed with an aim to monitor real time presence of alpha radioactive aerosols emitted from thorium nitrate hydrate. The results shows that the fabricated sensor can detect 0.0157 kBq to 0.1572 kBq of radio activity by radioactive aerosols generated from ($Th(NO_3)_4{\cdot}5H_2O$) at 0.1 ml/min flow rate. The proposed instrument will be helpful to monitor radioactive aerosols in/around a nuclear facility, building construction sites, mines, and granite polishing factories. © *2015 AIP Publishing LLC.* [http://dx.doi.org/10.1063/1.4922463]

The earthquake and tsunami in northeastern Japan led to a series of accidents at "Fukushima-1" nuclear power plant due to the failure of reactors' cooling systems, causing discharge of radioactive products. It is reported that, accidents at nuclear power plants, only gaseous fission products leak into environment, as well as nuclides having volatile precursors in the isobaric decay chains and formed from them directly into the atmosphere.[1,2] The workers involved in the processes employed to obtain concentrates of monazite can inhale airborne particles containing thorium, uranium, and other elements.[3] Previous researches show the feasibility of thoron detection (with mean thoron concentration of 9.6 kBq m^{-3}) and thoron decay products measurements[4,5] in an occupied indoor environment; however, they are bulky and need calibration, trainings to use them. Hence, there is a need to develop easy to use, compact radioactive aerosol monitor at nuclear power plants and domestic applications such as building construction sites, mines, and granite polishing factories.

In general, offline scintillation counting or alpha spectrometry is employed to track the alpha emitting radionuclides like uranium and thorium in the environment of a nuclear facility. Solid State Nuclear Track Detectors (SSNTD's) offer an alternative technique to measure radionuclides at ultra-trace level in the environment.[6–8] In the category of SSNTDs, Diethylene-glycol bis (allylcarbonate) (CR-39) is commonly used for monitoring alpha radiations.[9–12] However, SSNTDs unable to provide the radiation data instantaneously as after exposure for certain time duration, one has to carry out chemical etching and quantification of track density using optical microscope in the lab.[13,14] Hence, this technique is simple but it is off line, which involves time consuming chemical treatment and thus is not effective during any emergency situation requiring prompt response. Recently, our group has reported the first ever attempt to use reflectance based fiber optic sensor to monitor CR-39 detector exposed to alpha radiations emitted from 232Th source.[15,16]

From the continuation of our previous work, in the present study, effort has been made to develop real time fiber optic sensor utilizing CR-39 detector to monitor the radioactive aerosols generated from the thorium nitrate hydrate ($Th(NO_3)_4{\cdot}5H_2O$) for the first time. Analytical reagent grade powders of Th $(NO_3)_4{\cdot}5H_2O$ were procured from Sigma Aldrich, Korea and used as received. Stock solution of thorium was prepared by adding the above powder (1, 5 and 10 grams) in 25 ml of 0.1M HNO_3. The commercially available CR 39 films were used (500 μm) for this study (Kodak, USA). These films were cut in equal size of 10 mm × 10 mm to fit in the flow chamber assembly.

The lab designed flow chamber is shown in Figure 1(a) is used for the evaluation of radioactive aerosols generated from Th $(NO_3)_4{\cdot}5H_2O$. The CR 39 film is placed inside the flow chamber and the bifurcated OF (50:50, Industrial Fiber Optics, Inc., IF-562) probe is used to transmit the light and measure the reflected intensity from the surface of CR-39 film. The CR-39 film placed in a flow channel in such a way that the generated aerosols will interact with the film while passing through the flow channel. The overall dimensions of the flow chamber are 50 mm × 35 mm (H × W) and the volume of the flow chamber is 2750 mm^3. Two approaches were adopted to measure the reflected intensity from CR-39 film as described in Figure 1(b). In the first case, i.e., type A, the radioactive aerosols are passing through the fiber tip and CR-39 film, where as in type B, the radioactive aerosols are passing bellow the CR-39 film.

[a)]A. Kulkarni and S. Ha contributed equally to this work.
[b)]Authors to whom correspondence should be addressed. Electronic addresses: tkim@skku.edu and moonsoo@skku.edu

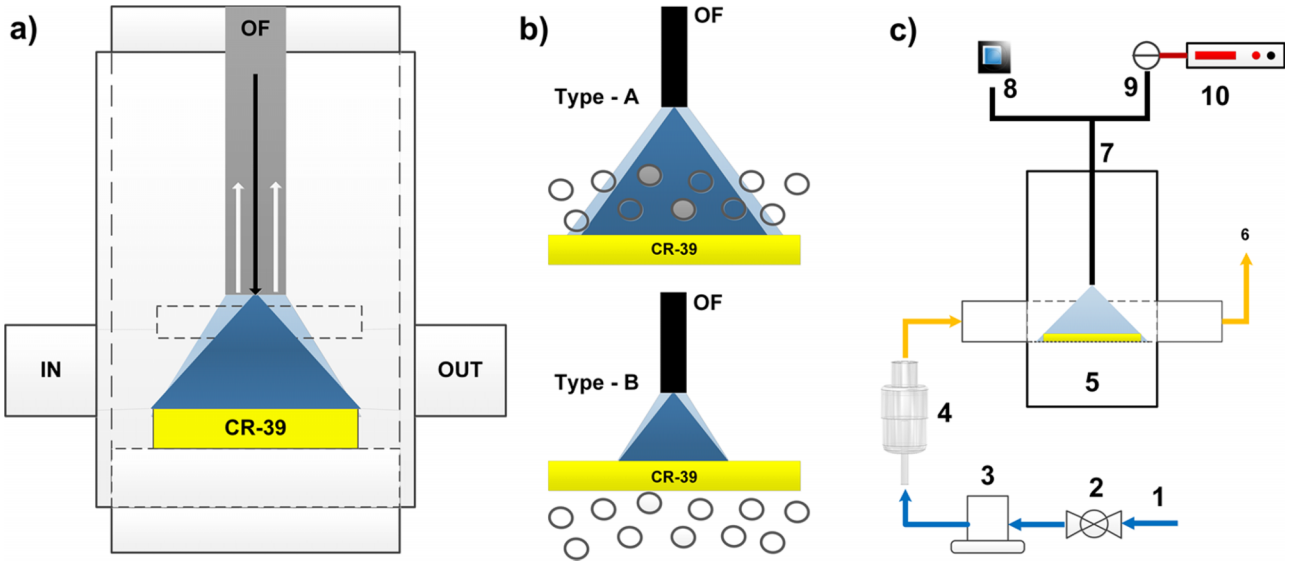


FIG. 1. (a) Schematic of lab designed radioactive aerosols flow chamber with CR-39 film and optical fiber (OF) placement for reflected intensity measurements. (b) CR-39 film placement in the flow chamber, in Type-A configuration the radioactive aerosols flowing through the film and optical fiber, where as in Type-B configuration the radioactive aerosols flowing bellow the CR-39 film. (c) Schematic of experimental setup for evaluation of effect of radioactive aerosols on CR-39 film. (1) Compressed air, (2) control valve, (3) Mass flow controller, (4) Nebulizer, (5) flow chamber, (6) vent, (7) bifurcated optical fiber probe, (8) light source, (9) photo detector, and (10) multimeter.

The setup used for the investigation of the interaction of CR-39 with radioactive aerosols is depicted in the Figure 1(c). Dry air (0.1 ml/min) is passed through the nebulizer using mass flow controller. The side stream reusable nebulizer (HS860, Philips Respironics, USA) is used to generate the radioactive aerosols from Th $(NO_3)_4{\cdot}5H_2O$ dissolved in 25 ml, 0.1M HNO3. The radioactive aerosols from nebulizer are then allowed to pass through the lab designed flow chamber and vented out safely in 3 stage scrubber containing 3M HNO_3. The light source (460 nm) (Newport, USA) is launched into the fiber, and the reflected light was measured using a Si photodiode detector (Thorlab, PDA36A). A digital multimeter (Keithley, 2700) was used to retrieve the sensor output and the data were recorded on a personal computer (PC), which were used to analyze the sensor performance. The generated radioactive aerosols were characterized for number concentration and size distribution using aerosol measuring setup similar to reported in our previous work.[17,18]

Initially, the CR-39 film configurations, viz., Type A and Type B (please refer Figure 1(b)) were exposed to Th $(NO_3)_4{\cdot}5H_2O$ radioactive aerosols for the duration of 800 seconds to monitor change in reflected intensity and track density. After exposure time, the CR-39 films were taken out and track density was evaluated after chemical etching. It is known that the radioactive aerosols causing much damage to surface morphology of the CR-39 film in short span of time. When the incident light interacts with the CR-39 film, a fraction of light is reflected from the surface (both top and bottom) and bulk, another fraction is absorbed, and the remainder is transmitted through. The microscopic images of CR-39 films for Type-A and Type-B configuration after exposure of radioactive aerosols are shown in Figure 2. After standard etching process, it is observed that the track density (number of tracks per unit area) in Type-A configuration is much higher (3000/cm^2) than that of Type-B configuration (138/cm^2). The interaction of radioactive aerosols with CR-39 film leads to chemical changes like chain scission/cross linking leading to permanent structural damage (tracks development) and hence there is a change in surface morphology. In Type-B configuration, the radioactive aerosols were interacted at the bottom of CR-39 film and the reflected light is collected from top surface. In this configuration, the surface morphology of the top surface of CR-39 film is mostly unaltered and hence less changes in the reflected intensity. It is interesting to note that Type-B configuration is similar to our previous report,[15] where the sensitivity of the sensor is better than in the present study. This may be due to the type of radiation type or source. In the earlier report, the CR-39 films were exposed to 232Th powder and in the present study CR-39 films exposed to radioactive aerosols generated from Th $(NO_3)_4{\cdot}5H_2O$ dissolved in 25ml, 0.1M HNO_3. However, in Type-A configuration, the CR-39 top surface is exposed to radioactive aerosols and the reflected light is also collected from the same surface where structural damages are more causing change in reflected intensity is higher than that of Type-B.

Further, the reflected intensity of CR-39 film was recorded online during exposure with Type-A configuration

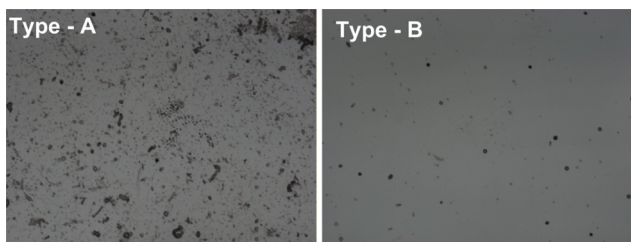


FIG. 2. Optical microscope image (before etching CR-39 film) for Type-A and Type-B sensor configuration after the exposure of radioactive aerosol.

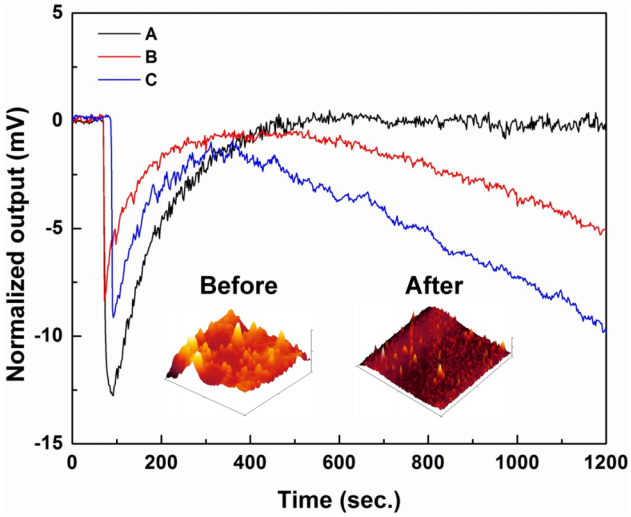


FIG. 3. Sensor response to radioactive aerosols from 232Th for various concentrations. Curves A, B, and C are 1, 5, and 10 gram Th $(NO_3)_4{\cdot}5H_2O$ in 25 ml of HnO_3, respectively (inset: AFM 3D image for before and after radioactive aerosol exposure for 10 gram Th $(NO_3)_4{\cdot}5H_2O$ in 25 ml of HnO_3).

for various concentrations of radioactive aerosols (1 gram, 5 gram and 10 gram of Th $(NO_3)_4{\cdot}5H_2O$ in 25 ml of HNO_3) and is as shown in Figure 3. The specific activity for Th $(NO_3)_4{\cdot}5H_2O$ is 3.93 kBq. Thus, the corresponding radio activity for 0.1 ml/min of flow rate is from 0.0157 kBq to 0.1572 kBq for used Th concentrations. The number concentration of Th radioactive aerosols for 10 gram of Th $(NO_3)_4{\cdot}5H_2O$ in 25 ml of HNO_3 is 202000 #/cc with mode particle size of 300 nm (data not shown here). The initial dip in the reflected intensity (at ~100 seconds) may be due to the sudden change in the flow chamber environment, i.e., air to radioactive aerosols. However, after about 100 seconds, the OF sensor starts to respond to the radioactive aerosols. It is observed that for 1gram Th $(NO_3)_4{\cdot}5H_2O$ in 25 ml of HnO_3, the reflected signal recovered to the normal, which implies the energy of radioactive aerosols for this concentration is not enough to change the morphology in the CR-39 film or it is beyond the detection limit of the photodetector used. However for other two concentrations, the reflected intensity changes significantly and continuous. These results suggest that the change in the reflectance intensity as an excellent means to monitor the presence of radioactive aerosols. It is well understood that the reflectance is strongly dependent on film surface topology, i.e., CR-39 film roughness. To characterize surface topology of CR-39 film, the surface roughness of irradiated films was measured using atomic force microscopy (AFM) in terms of the root mean square (RMS) roughness. Before exposure of radioactive aerosols, the CR-39 RMS roughness is 5.78 nm; however, after exposure to 1gram, 5gram, and 10 gram of Th $(NO_3)_4{\cdot}5H_2O$ in 25 ml of HNO_3, the observed RMS roughness is 2.55 nm, 2.44 nm, and 1.69 nm, respectively, which supports the reflected intensity (Figure 3) changes in accordance with the CR-39 film roughness. This finding is also interesting as the RMS roughness reduced the reflected intensity is also decreased, and it is opposite to our previous finding. This can be explained as, as the RMS roughness reduced, the multiple reflections form the film surface also reduced and hence the reflected intensity decreased continuously after exposure of radioactive aerosols.

In conclusion, the present work shows that the changes observed in CR-39 on exposure to alpha radioactive aerosols are significant with respect to the topological changes and optical reflectance intensity changes.

This work was partially supported by NRF under the Ministry of Science, ICT and Future Planning, Korea (NRF-2012M3A6A7054863) and GRRC program in Sungkyunkwan University.

[1]K. Hirose, J. Environ. Radioact. **111**, 13–17 (2012).
[2]Y. Kanai, J. Environ. Radioact. **111**, 33–37 (2012).
[3]K. D. da Cunha, J. A. M. Pereira, and C. V. B. Leite, Aerosol Sci. Technol. **32**, 453–464 (2000).
[4]J. Irlinger, M. Wielunski, and W. Ruhm, Rev. Sci. Instrum. **85**, 022106 (2014).
[5]S. Gierl, O. Meisenberg, T. Haninger, M. Wielunski, and J. Tschiersch, Rev. Sci. Instrum. **85**, 22013 (2014).
[6]R. L. Fleischer, P. B. Price, R. M. Walker, and E. L. Hubbard, Phys. Rev. **133**, A1443–A1449 (1964).
[7]M. F. Zaki and Y. H. El-Shaer, Pramana **69**, 567–574 (2007).
[8]A. R. Moorthy, C. J. Schopfer, and S. Banerjee, Anal. Chem. **60**, 857A–860A (1988).
[9]B. Center and F. H. Ruddy, Anal. Chem. **48**, 2135–2139 (1976).
[10]N. Dwaikat, M. El-hasan, M. Sueyasu, W. Kada, F. Sato, Y. Kato, G. Saffarini, and T. Iida, Nucl. Instrum. Methods Phys. Res., Sect. B **268**, 3351–3355 (2010).
[11]S. A. Durrani and R. K. Bull, in *Solid State Nuclear Track Detection: Principles, Methods, and Applications* (Pergamon Press, Great Britain, 1987), Vol. 111.
[12]J. W. Mitchell and S. Carpenter, Anal. Chem. **78**, 6955–6958 (2006).
[13]V. Chavan, P. C. Kalsi, and V. K. Manchanda, Nucl. Instrum. Methods Phys. Res., Sect. A **629**, 145–148 (2011).
[14]K. N. Yu, H. H. W. Lee, A. W. T. Wong, Y. L. Law, S. F. L. Cheung, D. Nikezic, and F. M. F. Ng, Nucl. Instrum. Methods Phys. Res., Sect. B **263**, 271–278 (2007).
[15]A. Kulkarni, C. K. Vyas, H. Kim, P. C. Kalsi, T. Kim, and V. Manchanda, Sens. Actuators, B **161**, 697–701 (2012).
[16]P. M. Joshirao, J. W. Shin, C. K. Vyas, A. Kulkarni, H. Kim, T. Kim, S. W. Hong, and V. K. Manchanda, Appl. Radiat. Isot. **81**, 184–189 (2013).
[17]A. Kulkarni, J. H. Lee, J. D. Nam, and T. Kim, Sens. Actuators, B **150**, 154–159 (2010).
[18]H. Kim, J. C. Yang, and T. Kim, Electrochem. Solid-State Lett. **13**, H137–H140 (2010).