A FLEXIBLE, SI-BASED NEUTRON DETECTOR FOR SAFEGUARDS APPLICATIONS

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ABSTRACT

Neutron detectors are useful tools for detecting radioactive material in safeguard and homeland security applications. Unlike photons that can be easily shielded, neutrons readily penetrate shielding material and have low natural background. With the proper application of neutron detectors useful information on radioactive materials can be obtained. Neutron detection is currently dominated by the use of compressed He-3 and BF-3 gas tubes. Field applications involving these types of neutron detectors can be problematic. For best detection results these detectors should be as close to the source as possible, but the geometry of these neutron tube detectors can be limiting. In addition, with the current shortage of He-3 and the hazardous material issues of BF-3, alternative technologies are being investigated. This talk will explore the possibility of integrating a layer transfer technique with neutron detection to fabricate a flexible neutron detection system. By employing these types of detectors, it is hoped a high detection efficiency will be realized by maximizing surface area (therefore, maximizing neutron interaction in the detecting medium) and having close proximity to reduce interference from other sources. Flexibility will assist in measurements occurring in tight configurations as well as optimize detectability in high background areas such as nuclear fuel cycle facilities. Details of the fabrication process, technological issues, and field application results will be discussed.

INTRODUCTION

This work is a result of current concerns regarding the shortage of viable helium-3 gas that is used for neutron detection purposes in the nuclear industry. Specifically, neutron detectors are essential to the nuclear safeguards and security cause in that most special nuclear material exhibits a common characteristic of emitting neutrons. For nuclear security concerns, merely the material's location and identification is needed, whereas, in safeguards, a quantifiable amount of neutron-emitting material must be surmised. The intent of this project is to ultimately quantify special nuclear material via gross neutron counting, but at the extent of this project thus far, a proof of concept and a prototype were initiated for developing a silicon-based neutron detector for use in safeguards.

JUSTIFICATION

A dominant method for determining the enrichment of ^{235}U in UF_6 is through passive gamma ray measurement. In these measurement scenarios, an inspector accumulates a spectrum at a given point on the cylinder with a high purity germanium detector (HPGe). Typically, the detector is collimated to reduce the influence of background radiation on the measurement. The counts (or count rate) from the 186-keV photon (region of interest for ^{235}U) and the 1001-keV photon (region of interest for ^{238}U) can be compared to determine the enrichment or mass of ^{235}U within a uranium hexafluoride (UF_6) cylinder. This, of course, requires the use of a large calibration standard or the use of an ISOCS-calibrated detector.[1] A disadvantage to using this type of photon measurement is the 186-keV gamma ray can be easily attenuated by the material in canister as well as the cylinder itself.

Figure 1. Accumulated UF6 Cylinders at an Enrichment Plant Site

Measuring the emitted neutrons from a UF_6 container can also be used to verify uranium enrichment. The advantage to using neutrons as a measurement of enrichment is that, unlike photons, neutrons are not easily shielded in this matrix. Neutron production rates for fuel-level enriched uranium in metallic form emit very few neutrons (———). However, in the chemical form of UF_6, neutrons are emitted in much greater numbers through the alpha-n reaction (———). In enriched uranium samples, ^{234}U is the dominant alpha emitter and the principle source of neutrons in UF_6. In the enrichment process, ^{234}U follows the enrichment of ^{235}U. This technique is only good for low enrichment (<5%) because the ratio of ^{235}U /^{234}U is relatively constant at these low enrichments. At higher enrichments, the isotopic concentration of ^{234}U should be known to determine accurate estimates of enrichment.[2]

A NEW DETECTOR

As aforementioned, there are several systems used in the field today for quantifying special nuclear material by analyzing emitted radiation in the form of photons, neutrons, neutrinos, and alpha and beta particles. For the application of concern to the investigators, a reliable and alternative method to measuring gross neutron counts from an unknown sample of special nuclear material (SNM) has been proven in theory. The SNM of interest is low enriched uranium (LEU) which spontaneously emits neutrons from the uranium-238 isotope (the major constituent of the uranium). This characteristic is to be used to assess material of unknown quantities in enclosed storage containers or cylinders housed throughout nuclear fuel cycle facilities around the world. There have been several studies to the feasibility of quantifying material in a non-destructive manner by several entities including U.S. national laboratories and the International Atomic Energy Agency (IAEA).[3] Studied technologies include energy spectrum counting from gamma-ray radiation, analyzing the energies from alpha radiation, and assessing neutron emissions. Past detectors for LEU have focused on scintillator- and semiconductor-based technologies for counting photon interactions: sodium-iodide crystals, cadmium-zinc-telluride crystals, lanthanum-halide crystals, and high purity germanium crystals.

The detector system in development at TAMU is a silicon-based neutron detector using ultra-thin wafer technology to create a large, flexible neutron detector that would ultimately be used as a blanket to cover the sample for maximized coverage and high volume count rates. A system such as this could easily match the curvature of any enclosure and ensure close to 4-π geometry than older neutron tube (or slab) detector technology.

POTENTIAL APPLICATIONS

As conveyed in Figure 1, a flexible, silicon-based neutron detector that can match the contours of a given container of neutron-emitting material could serve a substantial purpose in material verification measurements by inspectors to front-end and back-end nuclear fuel cycle facilities such as enrichment plants and reprocessing plants. Gamma spectrometry for material quantification is currently used in these environments but, due to the nature of photon interactions, data can be corrupted or clouded to give off-normal results (either benign or malicious). The added benefit of measuring neutron emissions from such SNM, verifies the gamma spectrometry data but also can acquire information in adverse environments where photon emissions are not adequate for precise measurements or analysis. Moreover, in bulk processing facilities, where SNM can flow through fixtures in various geometries, a flexible neutron counter can be of great benefit for qualifying the processes material.

For future use, the notion of a flexible, silicon-based neutron detector can be beneficial to other industries/applications beyond safeguards: permanently fixated process monitoring for operators or intelligence gathering by wearing effective neutron detectors as garments while visiting a facility suspected of undeclared activities. The expected use of this detector can benefit various areas within the nuclear industry.

DETECTOR DEVELOPMENT

Researchers at TAMU are working on the determining the feasibility of developing a flexible silicon (Si) based neutron detector by using a wet etching-based layer transfer technique. In our demonstration, a 320nm-thick silicon layer was transferred onto a flexible polymer substrate by preferential etching of the buried SiO_2 layer in a silicon-on-insulator (SOI) wafer which was bonded with the polymer. Potentially, metal coating on both sides of the transferred layer will create a flexible neutron detection system. By employing such type detectors, it is hoped that a high detection efficiency will be realized by maximizing surface area (therefore, maximizing neutron interaction in the detecting medium) and, when in tight configurations, using the flexibility of such a detector to optimize detectability in high background areas such as nuclear fuel cycle facilities.

BACKGROUND

In 1977, W.K. Chu found that hydrogen implantation into Si can lead to exfoliation of the Si layer after thermal treatment.[4] Almost 20 years later, researchers realized the potential significance of this phenomenon in terms of a promising application: a new layer splitting process – the “Smart-cut.”[5,6] The process includes (1) a high dosage of hydrogen ion implantation, (2) wafer bonding to a handle wafer, and (3) annealing of the jointed pair. Under enough thermal budget, a complete shearing (or exfoliation) at the depth of hydrogen implants occurs which lead to the transfer of the Si layer onto another substrate. This layer transfer process has greatly impacted microelectronic device fabrication by becoming a mainstream technique to fabricate SOI wafers. Transistors fabricated by using SOI wafers are much faster. Furthermore, the buried insulation layers can also be used to enhance devices’ stabilities in harsh environments involving particle irradiation or high temperatures.

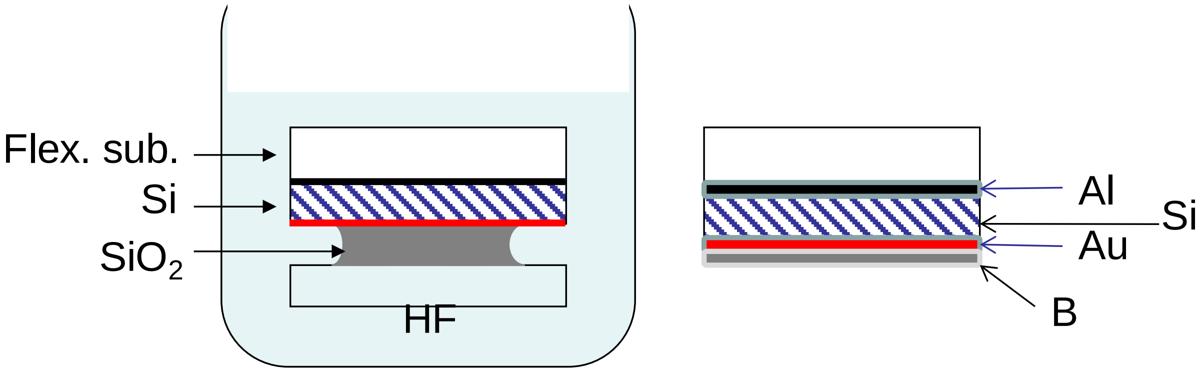


Figure 2. Schematics of the etching process to transfer a Si layer onto a flexible substrate

The Smart-cut technique is used to transfer thin layers onto another rigid surface. Transferring a silicon layer onto a flexible substrate to form a bendable device, however, is much more challenging (shown in Fig. 2). One issue is that a flexible substrate such as a polymer cannot survive high temperature annealing which is required to initialize hydrogen induced cracking in Si (>~500 °C). Thus, development of new layer transfer techniques requiring a low thermal budget is greatly needed. One solution is to wet etch a SOI wafer. As shown in Figure 2, for a polydimethylsiloxane (PDMS) substrate bonded with an SOI wafer, the Si membranes can be released and transferred onto the plastic substrate if the buried SiO_2 layer can be preferentially etched by dipping the structure into a hydrofluoric acid solution. With this, the feasibility of the approach to form a flexible radiation detection sheet has been proven.

Once the capability of manufacturing the material was proven, the project investigators started with a commercial SOI wafer purchased from a private vendor (SOITECH Inc). The wafer was manufactured by using the Smart-Cut technique. The top Si layer was n-type doped (20-50 Ohm cm) and <100> oriented with a thickness of 320 nm. The buried SiO_2 layer is about 3 microns thick. Rutherford backscattering spectrometry was used to characterize the sample. The normalized channeling yield of the surface peak was ~3%, which suggests a good crystalline quality of the top Si layer. The sample was cleaned and then was subjected to dehydration baking, spin coating of the photoresist, UV exposure, developing, Si etching, bonding with a polymer, dipping into hydrofluoric acid for SiO_2 etching, Si layer transfer, and metal coating. An optical microscope and a scanning electron microscope were used to characterize the structures after each fabrication step.

PROCEDURE

First, spin coating of AZ514 was used to produce a constant thickness of photoresist across the sample. Then prebaking at 110 °C for 1 minute was used to increase the sensitivity of photoresist to UV light. Samples were exposed to UV light by using Karl Suss MA-6 Mask Aligner under a power density of 7.2 mw/cm^2, followed by baking at 120 °C for 3 minutes. The exposure to MA6 was repeated under a power of 399 mw, followed by cleaning in acetone for about 20 seconds.

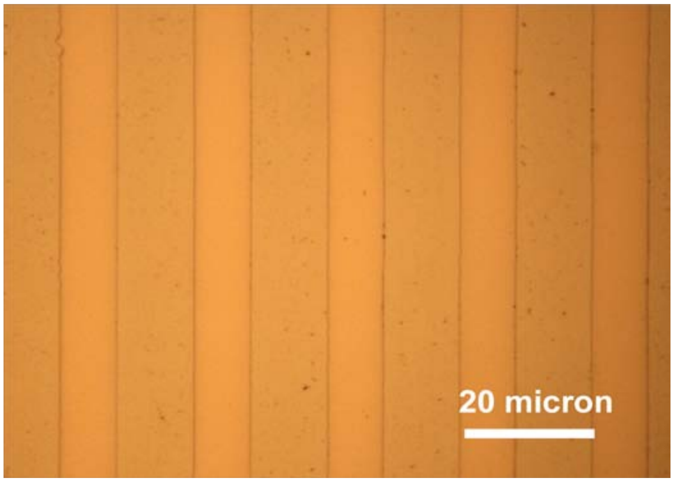
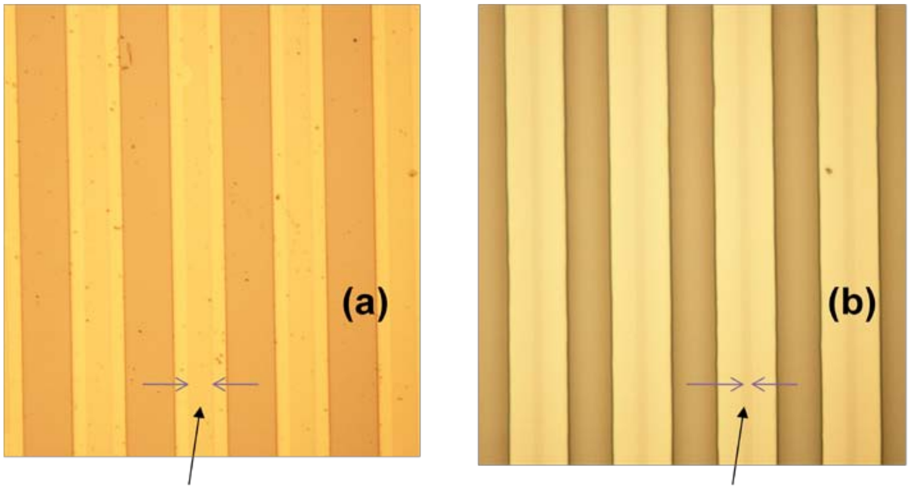


Figure 3. Optical image of a patterned SOI wafer with periodical stripes of 10 micron width



SiO_2, 1 min etching

SiO_2, 3 min etching

Figure 4. Optical images of etched samples in HF for 1 minute (a) and for 3 minutes (b)

Silicon etching was realized by using 20% KOH at 80°C for about 1.5 minutes. KOH etching is one the most commonly used methods to clean and etch silicon and the Si dissolution rate is well controllable. For the Si regions uncovered by photoresist, they are etched to the depth of the buried SiO_2 layer. Then, the buried SiO_2 layer was etched by dipping the structure into 49% HF.

Figures 3 and 4a show the optical image of the structure before and after etching in HF for about 1 minute. The layer transfer can be realized even though the SiO_2 etching was not complete. The arrows in Figures 4a and 4b correspond to the region with residual SiO_2 left since etching times were not long enough. Figure 4b shows the optical image of the HF-etched structured for an etching period of 3 minutes. With increasing etching times, the thickness of the residual SiO_2 was reduced down to ~1 micron only. The etching rate is about ~2.5 micron/min, but this rate depends on the width of Si stripes.

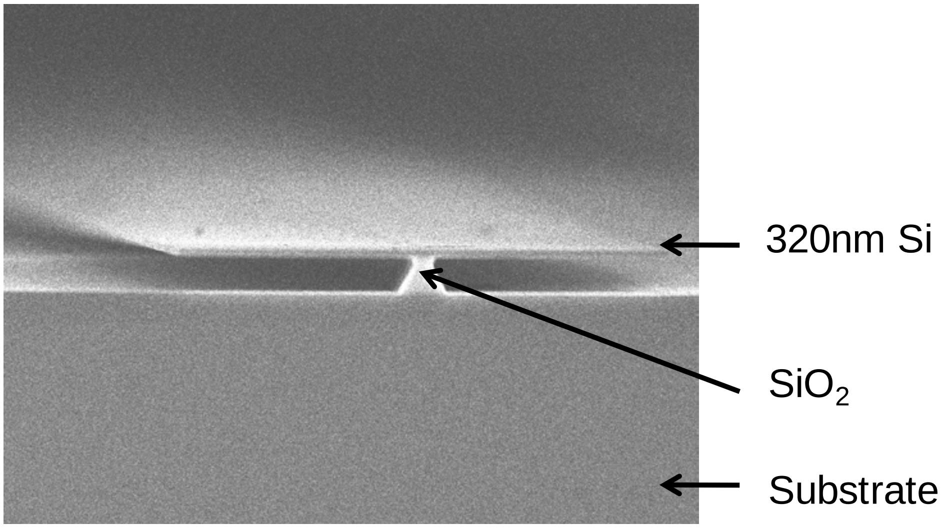


Figure 5. SEM image of the sample after HF etching

Figure 5 shows the cross sectional view of a cleaved structure obtained by using a scanning electron microscope. In order to make the top Si layer visible, the sample has an incomplete SiO_2 etching, so the residual SiO_2 can support the top Si layer.

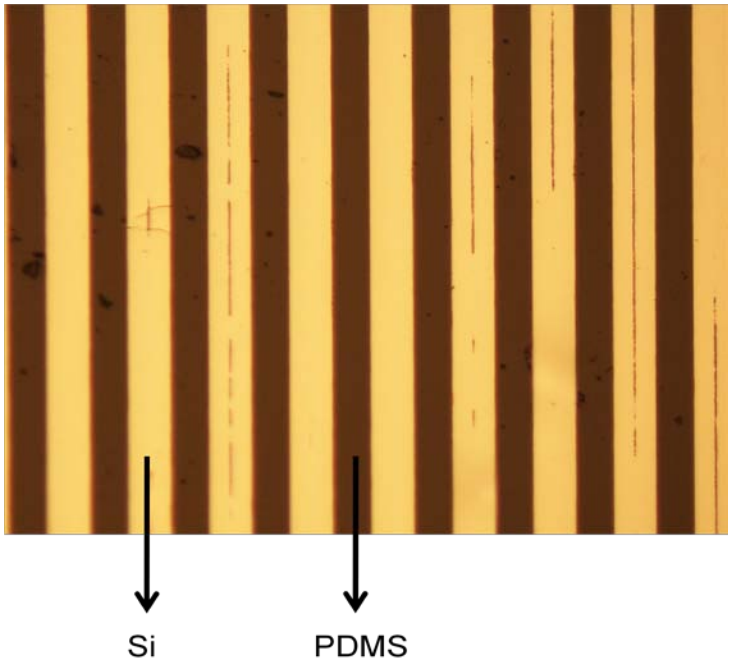


Figure 6. Optical image of the structure with Si stripes bonded with PDMS substrate

Figure 6 shows the structure transferred onto PDMS which is a silicon-based organic polymer that has very flexible polymer backbones. A thick rubber semi-solid of PDMS can be used as flexible substrate for the proposed device. The dark region in the figure corresponds to the PDMS substrate. The residual SiO_2 is visible as a narrow line, which can be easily removed by HF.

DEVELOPMENT RESULTS

The study so far has shown the feasibility to transfer a thin Si layer onto a flexible PDMS substrate. If gold (Au) and aluminum (Al) metal films are deposited before and after HF etching, the transferred structure will contain a Au/Si/Al multilayer. For the structure shown in Fig. 6, an Au layer has been buried between the Si layer and the PDMS substrate. This structure can be used as a Schottky barrier radiation detector (SBD). The SBD is one of the most widely used radiation detectors. For n-type Si having a Fermi energy higher than that in Au, electron movements into Au creates a depleted region in Si. Any carriers created in this region due to irradiation will be forced to move out under an internal electric field thus contributing electronic signals in the circuit. For safeguard applications, neutron detection can be realized by absorptive reaction $^{10}B(n,\alpha)^{7}Li$ with boron atoms coated on the top of the Au layer. One drawback of the design is that the thin body of Si strips is not thick enough to completely absorb reaction products. The majority of particles will penetrate through the Si body if the projected range of particles is much larger than the Si layer thickness. This makes it difficult to quantitatively measure the particles’ total energies.

If the silicon layers are isolated into small detection units, 2-dimentional mapping of neutron flux is possible. This will greatly benefit safeguard applications with the requirement of directional detection. Signal collection from individual units is accomplished relatively easily by integrating with well-established techniques for microelectronic device fabrication. Another advantage of the Schottky barrier detectors is that the required voltage bias is small (<100 V), which makes it possible to develop battery-driven portable detection systems. Currently, the system must be optimized to maximize flexibility and sensitivity: the width and thickness of the Si stripes must be optimized to avoid mechanical failure during bending. The Si doping level needs to be carefully controlled since the width of depleted region should be less than the Si layer thickness.

DETECTOR SUMMARY

Overall, the feasibility of transferring Si strips (230 nm thick) onto flexible polymer substrate has been adequately proven. The technique is based on bonding a SOI wafer with a PDMS substrate. With appropriate lithography and Si etching, trenches were developed and penetrated into the buried SiO_2 layer, thus providing paths for HF solution to etch SiO_2 so that the layer can be lifted off and transferred onto the PDMS substrate. With Au and Al metal film deposition on each side of the Si layer to form an Au/Si/Al Schottky barrier, the structure can be used to detect energetic particles such as helium particles created by neutron reactions with boron atoms. The boron atoms can be deposited on the top of Au layer. The structure can be developed into flexible 2-D arrays for two dimensional mapping of neutron flux for various safeguard applications.

CONCLUSION

With the positive results received from the previous section, a new detector has been proven to be manufactured (at least as a prototype). Further characterizing and testing is necessary whence the Si-based detector is complete but initial reports from the TAMU Electrical Engineering Department show promise. When available, the detector will be assessed for neutron counting capabilities and, if results are adequate, the detector will be considered for field testing. The main concern for the NUEN staff at TAMU is for determining the efficiency of such a detector with such thin layers of active material. A major concern is the penetrability of thermal neutrons and how the response of such a flexible detector could be affected. Without mention, further testing is needed but, thus far, the TAMU team has determined that the neutron detector system can be constructed. The subsequent step is for analyzing the detector's usability for counting neutron interactions.

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