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Flexible silicon-based alpha-particle detector

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The detection of alpha particles in the field can be challenging due to their short range in air of often only a few centimeters or less. This short range is a particular issue for measuring radiation inside contaminated pipework in the nuclear industry, for which there is currently no simple method available without cutting the pipes open. Here, we propose a novel approach based on a flexible 30 x 10 mm^2 sheet of 50 µm thin crystalline silicon. Following established fabrication steps of pn-junction diodes, we have successfully constructed a device with a signal-to-noise of >20 in response to 5.5 MeV alpha-particles using a bespoke amplifier circuit. As flexible detectors may readily conform to a curved surface and are able to adapt to the curvature of a given pipeline, our prototype device stands out as a viable solution for nuclear decommissioning and related applications.

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The decommissioning of nuclear sites requires reliable techniques for the detection, identification and quantification of radioactive substances. Discriminating between disposable and contaminated waste has a significant impact on the cost of decommissioning. The UK government has already forecast the enormous cost of at least £117 billion for decommissioning UK nuclear sites over the next century,[1] with other nuclear nations facing similar issues. New technologies and solutions therefore need to be explored in order to reduce the cost and to speed up the decommissioning progress.

In particular, there is no simple, low-cost solution available for assessing the presence of radioactive sources that are essentially alpha-only emitters, such as plutonium ^{239}Pu, on the inner walls of pipework. The range of alpha-particles in media is intrinsically short and as such, alpha-emitters are undetectable from the outside, as any radiation occurring on the inside of the pipe is absorbed by the wall. The emitting isotope may even be embedded in tiny cracks in the pipe wall meaning that alpha-particles may lose energy as they emerge. Consequently, alpha-particle detectors need to be positioned as closely as possible to the inner walls in order to achieve the greatest detection efficiency and be able to detect very low radiation doses.

For the detection of alpha-particles, one can either collect the generated free charge carriers (formed directly by ionization of atoms or molecules) as an electrical pulse or convert the scintillation photons (due to recombination of free charge carrier pairs) into an electronic signal.[2]

One commercialized technique is the Ionsens® pipe monitor[3] which is based on the measurement of air-driving ions: if ambient air is struck by alpha-particles, the ionized air molecules can be driven to an ionization chamber via a fan. Collecting the ionized molecules and modelling the airspeed distribution inside the pipe allows one to determine the drifted distance from the source to the chamber, and so to locate the alpha-radiation.[4] However, as this method requires a testing chamber for inspection, each pipe must be cut into segments – which is incompatible with the inspection of extended pipework.

Other commercial methods, such as the Alpha Explorer™ and Pipe Crawler®,[5-7] are based on the scintillator detector ZnS(Ag) which is most extensively used in nuclear facilities and the like:[8] when struck by an alpha-particle, the scintillator absorbs its energy and re-emits the absorbed energy as photons, revealing the presence of alpha-emitting isotopes. However, these methods would have a low detection efficiency of 1%,[7] a low energy resolution and may be sensitive to other ionizing radiation in the environment as well as local alpha radiation.

Here, we suggest a novel detector system based on flexible crystalline silicon that can conform to the inner diameter of pipework for the direct detection of alpha-particles.[9] Our solution not only offers the potential for 360-degree inner diameter coverage, but it can also be adapted to pipes of different diameter. By being as close as possible to the inner pipe walls, maximum sensitivity and uniform efficiency can be achieved, even for low energy alpha-particles, where other approaches may struggle. As a proof-of-principle, we demonstrate that a sheet of flexible crystalline silicon of 50 μm thickness, as shown in Fig. 1, forms the basis for a high-performance in-pipe alpha-particle detector. Our sensor can be more generally adapted to any curved surface, such as the exterior of barrels filled with radioactive waste. It can also address the requirements of some nuclear and particle physics experiments that require non-planar detectors.

FIG. 1. Crystalline silicon wafers become flexible at ca. 70 µm thickness. Accordingly, they can be bent to be used for screening the inner surface of a 2” diameter pipe for alpha-contamination and can be mounted on an elastic inspection gauge.

For nuclear radiation detectors, high-purity float zone wafers are particularly suitable because of their high resistivity and low defect density.[2] However, we opted for Czochralski-grown wafers, because of their inherently high oxygen content. Oxygen not only strengthens the material (reducing wafer breakage during fabrication), but is also helps withstand the effects of radiation damage by alpha-particles.[2] In addition, Czochralski wafers are lower cost than float-zone wafers. We then introduce phosphorus doping by thermal diffusion using a solid planar diffusion source (PH-1025, Saint-Gobain). Phosphorus and p-type wafers were chosen, because boron as dopant source for n-type wafers would have required additional processing steps for the removal of the borosilicate glass layer (also referred to as boron skin), and hence would have increased the risk factors for wafer breakages during processing. The source wafer is placed immediately adjacent and parallel to the 3” p-type silicon wafers and the doping step is carried out at 1025 °C in an N_2 atmosphere for 60 minutes. After unloading, the excess glass which forms during the doping process is removed from the silicon wafers by HF processing. This is followed by the deposition of a 200 nm thick aluminum layer on both sides of the wafer (MANTIS HEX® thermal evaporation system). The coating serves both as an electrical contact and as an optical blocking filter to remove daylight sensitivity of the produced detector. Using Monte-Carlo type simulations (GEANT4®, Fig. 2),[10] we have determined that the energy loss of alpha-particles is negligible for this thickness of aluminum, which in turn completely blocks optical photons, thereby minimizing background noise.

The fabrication process requires additional steps to improve the detector's performance. These include thermally growing, a 500 nm thick layer of sacrificial oxide on the wafers before starting the doping process, in order to remove damaged surface regions and contamination.[11] The thermal oxide is also used as a doping barrier on the back side of the wafer in order to ensure that only a single pn-junction is formed. A film of organic lacquer (Lacomite® varnish) was used to protect the oxide when opening the window for doping. Lacomite® is a cheap, air-drying and acetone-soluble resin. Finally, RCA cleans were performed prior to each high-temperature processing step.

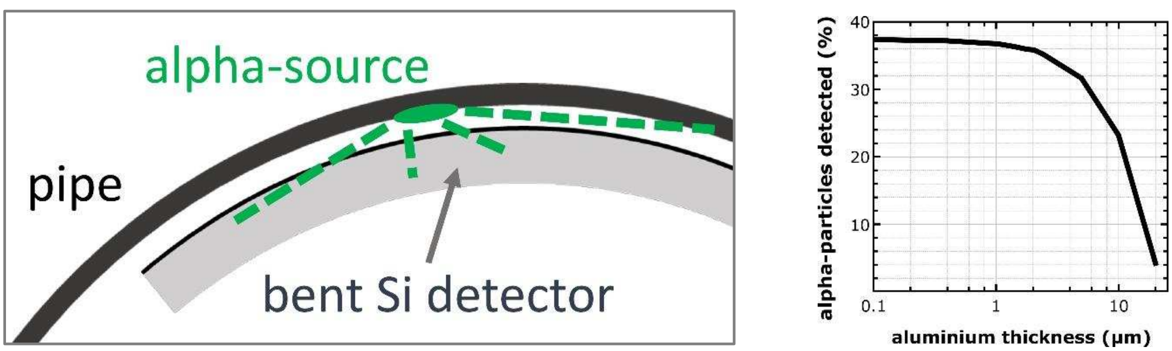

FIG. 2. GEANT4 model for a Si detector in an alpha-contaminated steel pipe (left hand side).[10] Alpha-particle tracks in air are indicated as green dashed lines. The model was created to understand the effect of Si contact material and thickness as well as the effect on detection efficiency from air gap between detector and ^{239}Pu. If the aluminum on the top face of the detector is less than 1 μm thick, it does not significantly affect the detection efficiency, because the energy loss of alpha-particles remains negligible, as shown in the graph on the right. While 50% of alpha-particles are absorbed by the pipe wall, 10% will miss or reach the detector at a grazing angle, such that only 40% of alpha-particles remain detectable at best.

Alpha-particles from ^{239}Pu have a typical kinetic energy of 5.1 MeV, corresponding to a Bragg peak around 22 µm deep in silicon.[12] Therefore, the vast majority of free charge carriers will be created within the first 20 µm by the ionization of the silicon atoms and so the 50 µm thickness of the wafer is sufficient to fully absorb even the highest energy alpha-particles. At the same time, the low wafer thickness and low background doping (8×10^{14} cm^{-3}) ensures that the generated electron-hole pairs can easily diffuse to the depletion layer and generate a current before recombining.[13] Hence, while the 50 µm thickness was initially chosen for its mechanical flexibility, it is also favorable for alpha-detector operation.

We determined a pn-junction depth of ca. 2.5 µm via a thickness measurement of the phosphosilicate glass on the doped wafers. The junction depth is then read-off from the datasheet of the planar diffusion source (PH-1025, Saint Gobain). Here, a glass layer of 100 nm was found using a bespoke ellipsometer.

Since the doping concentrations largely differ from each other, the depletion region of the pn-junction extends predominantly into the low doped side and can thus be determined by the resistivity ρ of the substrate wafers. Here, using a four-point probe measurement we found a sheet resistance of 4 kΩ/□ and hence a resistivity of 4 kΩ × 50 µm = 20 Ωcm. Assuming a built-in voltage of $V_b \approx 0.5$ V and a typical hole mobility of 400 cm^2/Vs in Si, with the dielectric constant $\varepsilon \approx 1$ pF/cm, the depletion width w becomes[2]

$$w = \sqrt{2\varepsilon\mu\rho V_b} \approx \sqrt{2 \text{ pF cm}^{-1} \times 4\,000 \text{ cm}^3 \text{ F}^{-1}} \approx 0.9 \text{ µm}. \tag{1}$$

On the other hand, the depletion layer thickness can also be estimated from the measured capacitance of 10 nF/cm^2,

$$w = \varepsilon \frac{A}{C} \approx 1 \text{ pF/cm} \times \frac{1 \text{ cm}^2}{10\,000 \text{ pF}} = 1 \text{ µm}, \tag{2}$$

which agrees well with the above calculation.

Multiple 30 x 10 mm^2 samples were then cleaved from flexible silicon wafers with different background doping levels (as discussed in the following) and mounted onto a 2” pipeline inspection gauge. The current-voltage characteristics of the mounted devices confirm the excellent pn-junction characteristics as shown in Fig. 3.

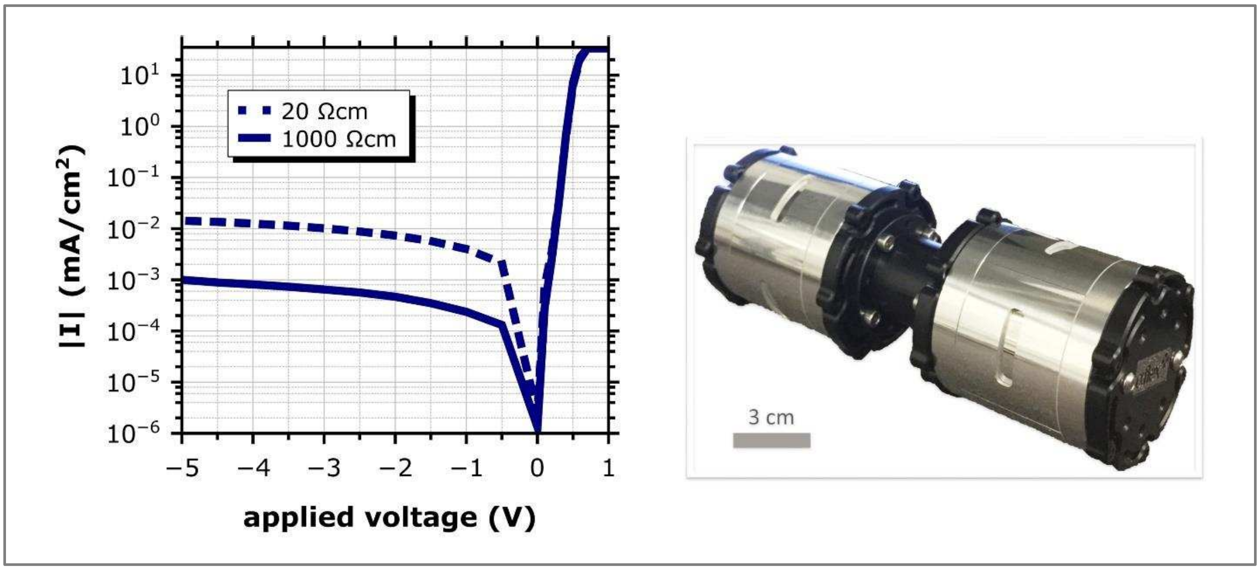

FIG. 3. The electrical current density I (in mA/cm^2) as a function of the applied voltage (in V) is shown on a log-scale (left figure) for samples of 3 cm^2 in size, which were mounted onto a 2” pipeline inspection gauge consisting of two modules (right figure); the compliance limit was set to 100 mA. Here, the dotted and solid curve represent detectors fabricated using a 20 and 1 000 Ωcm resistive substrate, respectively. The dark currents range from less than 1 nA/cm^2 at 0 V to a few μA/cm^2 at 5 V. The flat reverse current along with the sharp onset at 0.5 V forward bias indicates the high-quality of the pn-junctions. We anticipate from our GEANT4 simulations that four modules can cover 20 m length of pipe in ca. 60 min, with a minimum detectable activity of 0.4 Bq/cm^2.

The initial alpha-particle detection experiments were conducted with the 20 Ωcm material. Given the relatively large detector size and typical capacitance of almost 30 nF, matching an amplifier to the large capacitance of the device was essential. Nevertheless, by using a bespoke charge sensitive amplifier – made from off the shelf electronic components (to reduce overall costs) – 5.5 MeV alpha-emission of a ^{241}Am source (40 kBq) from 3 mm distance was detected with a signal-to-noise ratio of 4; the distance was chosen for mechanical and operational reasons such as roughness and welding joints. The signal-to-noise ratio generally increases with decreasing capacitance, it is important to keep the capacitance low. To this end, we introduced higher resistivity (1 000 Ωcm) silicon material. Based on Poisson's equation, the depletion width of a pn-junction scales with the square root of the resistivity (see Eq. 1), so increasing the resistivity of the wafer should increase the depletion width and decrease the capacitance of the device (see Eq. 2). Fig. 4 demonstrates this benefit experimentally. The measured device capacitance has dropped from almost 10 nF/cm^2 to 2 nF/cm^2. We also obtained an improvement of the signal-to-noise ratio from 4 to more than 20, as shown in the inset of Fig. 4.

Finally, since beta and gamma particles could influence the detection of alpha radiation, we also simulated the response to beta and gamma rays at various energies. While the gamma signal generally scales with the silicon layer thickness or junction depletion width, we found that a 50 µm thin and flexible silicon detector is very inefficient for such type of rays when operated without bias (photovoltaic mode); given it is so then it is ideal to be used in a high background environment. In fact, we could not observe any gamma signal, when we tested our detector to a high gamma source, such as ^{60}Co, which is a common activation product in aged nuclear waste.

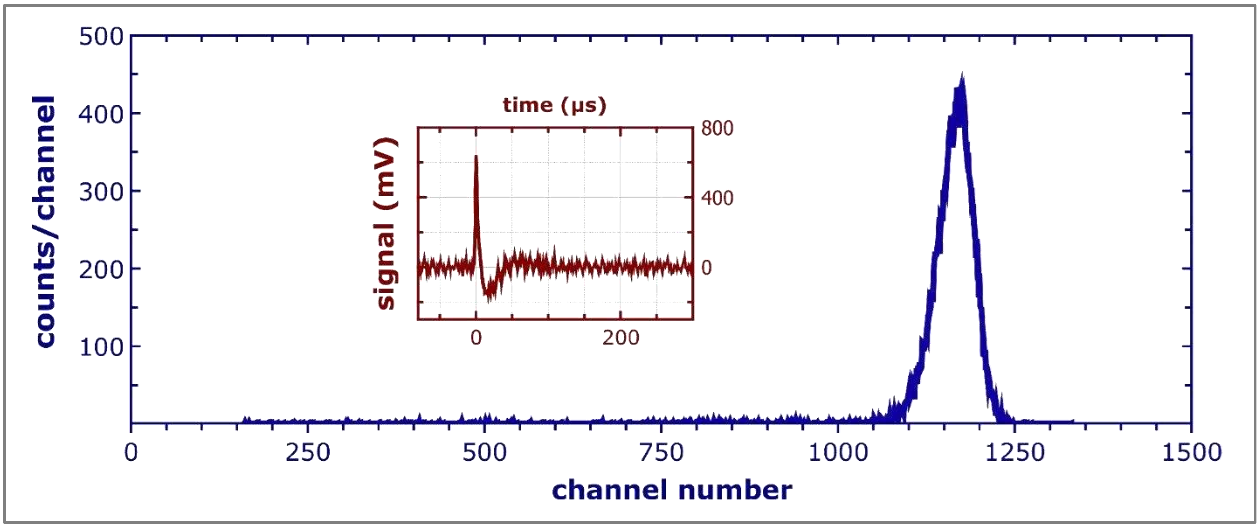

FIG. 4. The spectral response of a 30 x 10 mm^2 un-biased, flexible silicon detector (in photovoltaic operation and mounted onto a 2" pipeline inspection gauge) to 5.5 MeV alpha-particles of a ^{241}Am source with 40 kBq activity, using a bespoke charge sensitive amplifier circuit, a low-level discriminator (to reject pulses below 0.7 MeV) and an integration time of 10 min. No shifts in the peak position where observed over a three-week period, highlighting excellent repeatability and stability of the sensor system with a FWHM energy resolution of less than 5%. The signal peaks at 640 mV, compared to a noise floor of 30 mV, resulting in a signal-to-noise ratio of more than 20 (see inset). The devices were left unbiased for simplicity of design and cost reduction in the amplifier circuit

Overall, we have demonstrated the use of flexible silicon as a platform for radioactive particle detection. While flexible silicon has already found its way into photonics applications, e.g. solar cells,[14] it has not yet been used for applications in the nuclear industry, or for other scopes.

Flexible silicon could also be used for the efficient detection of thermal neutrons. Since the interaction of thermal neutrons with boron (^{10}B) leads to the emission of alpha-particles, a 2 μm thin layer of ^{10}B enriched oxide may act as the intrinsic region between two flexible silicon sheets of different doping types.[15] In principle, as such flexible pin-devices have a high thermal neutron efficiency, their applications could include the detection of thermal neutrons in different industrial scenarios.

Furthermore, in some nuclear and particle physics experiments, angle-resolved information needs to be collected. For example, a large-area, flexible, silicon detector could become the preferential alternative for setups, where the required cylindrical geometry has been approximated by a hexagonal cross-section.[16] Two-dimensional position sensing could be achieved by subdividing the outer electrode into pixels. We will focus on the potentials of a flexible sheet of silicon for nuclear and particle physics experiments in a future paper.

In summary, we have introduced and demonstrated a high-performance and low-cost solution for a novel alpha-particle detector based on flexible crystalline silicon, demonstrating that flexible silicon technology may have an interesting role to play in the field of nuclear decommissioning and related applications.

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[1]See https://www.gov.uk/government/publications/nuclear-provision-explaining-the-cost-of-cleaning-up-britains-nuclear-legacy for more information about the cost of decommissioning Britain's nuclear legacy; accessed 15 June 2017.

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