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Technology development for soft X-ray spectroscopy☆

P.T. Törmä *, H.J. Sipilä, T. Koskinen, M. Mattila

HS foils Oy, Tietotie 3, 02150 Espoo, Finland

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ABSTRACT

X-ray spectroscopy instruments lose part of their performance due to the lack of suitable components for soft X-ray region below 1 keV. Therefore, in the analysis of low atomic number elements including lithium, beryllium, boron and carbon instrument sensitivity is often limited. In this work we describe how the performance of the spectroscopy of soft X-rays is significantly improved when all devices integrated in the spectroscopic instrument are suitable for both soft and hard X-rays. This concept is based on utilizing ultra-thin SiN X-ray windows with proven performance not only as a detector window but also as an X-ray source window. By including a soft-X-ray-sensitive silicon drift detector with efficient surface charge collection in this concept the sensitivity and performance of the instrument is significantly increased.

1. Introduction

The energy range from 50 eV to 500 eV has been challenging for X-ray spectroscopy. X-ray detectors and X-ray sources have been technologically very demanding. Recently silicon nitride (SiN) windows [1–2] have opened new possibilities. Windows with thicknesses starting from 20 nm are vacuum tight and can be used for sealed detectors like proportional counters and silicon drift detectors (SDD). The transmittance of the ultra-thin SiN window for, e.g., Li Kα is about 50%, and, thus, it is clear that also photons with low energies from the measured target can pass through the window into the detector.

In commercial X-ray tubes the thinnest window typically consists of 50 μm of beryllium. Because silicon nitride foils tolerate high temperatures and are radiation hard, they can be utilized also as windows for X-ray tubes. This will significantly improve the soft X-ray excitation efficiency. For example, the signal from carbon, oxygen, and sodium will increase significantly under the same operational conditions of the X-ray tube if only the tube window is changed from a beryllium window to an ultra-thin SiN window.

Proportional counters have internal gain and it is relatively easy to measure very soft X-rays. Electronic noise does not cause limitations and practically all photons which pass through the window are detected. The situation is more complicated with SDDs. The detector surface may have dead layer which will cause additional absorption. This phenomenon has been studied [3–4] thoroughly for silicon solar panels. It is found that an Al_2O_3 layer deposited by the atomic layer deposition (ALD) method on silicon will cause negative charge on the surface and greatly improve charge collection. It is obvious that this is valid also on the SDD surface. In addition, this negative charge may improve the lifetime of the detector under a heavy radiation load by enhancing the charge collection.

In order to have the best soft X-ray performance the system needs to combine an X-ray source with a soft-X-ray-friendly exit window, an X-ray detector with a soft-X-ray-friendly entrance window and high soft X-ray sensitivity. In this work we show how the performance of the soft X-ray spectroscopy is improved compared to traditional instruments by the approaches mentioned above.

2. Methods

In this work we have characterized X-ray window structures having a 20-nm-thick SiN layer supported by a mesh. The freestanding SiN foil is covered by Al layers having a total thickness of 20 nm. The open freestanding window area is 79% of the total 20 mm^2 circular window area. The fabrication process of these SiN based X-ray window structures is described elsewhere [1]. Characterization results are compared to simulated and previously reported results from well known polymer based and novel graphenic carbon based X-ray windows [5–6].

The performance of the ultra-thin SiN X-ray windows against essential requirements has been extensively studied earlier [2]. Therefore, basic properties are not discussed in detail in this work. However, the 20-nm-thick SiN X-ray windows pass the basic tests as well as the windows in the previously reported work [2].

The radiation hardness of the 20-nm-thick SiN X-ray windows against high X-ray doses was proven with two X-ray sources. This was done for proving that these windows can be used in long lifetime

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* Corresponding author at: HS foils Ltd, Tietotie 3, 02150 Espoo, Finland.
E-mail address: pekka.torma@hsfoils.com (P.T. Törmä).

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operation in X-ray tubes and detectors. The first source was a laboratory X-ray tube running at 40 kV/40 mA. The samples were exposed to 8 keV X-ray radiation for 4 h. The tube had a 250 μm beryllium window and no further filtering was used. The second source was a white synchrotron having 2.5 GeV particle energy, a ring current of 120 mA, and a critical X-ray energy of 6 kV was used for high X-ray dose. The sample was exposed to a 6×2 mm^2 beam for 90 min. All the X-ray window samples were characterized before and after X-ray irradiation exposure. Characterization methods included 1.6 bar differential pressure cycle testing, He leak detection, optical microscopy and scanning electron microscopy. No visual changes were observed. Also there was no change in strength and the windows were He leak tight before and after the experiment.

The X-ray transmittance characterization was performed in the synchrotron radiation laboratory of the Physikalisch-Technische Bundesanstalt (PTB) at the storage ring BESSY II in Berlin, Germany [7]. The soft X-ray radiometry beamline [8] provided tunable monochromatized radiation in the spectral range from 50 eV to 1900 eV. A Hamamatsu GaAsP photodiode G1127–04 was used for radiation detection in the EUV reflectometer [9]. The spectral transmittance was measured by consecutive measurement of first the incoming photon beam, second the transmitted beam and third the incoming beam again to assure stability of the source. The transmittance was calculated by normalizing the second scan data with the reference scan data.

The transmittance spectra of the graphenic carbon and polymer X-ray windows were calculated with a self-made software based on previously published atomic scattering factor data [10].

A Monte Carlo simulation model was built in order to study the advantages of using a SiN window in an X-ray tube for low-Z fluorescence excitation. This model simulates a setup where the carbon concentration of a cast iron sample is measured using an X-ray tube and a silicon detector. The model was built using the Geant4 simulation toolkit [11] with PENELOPE physics processes. First, spectra for several X-ray tube targets were simulated. In each simulation a thick target was bombarded with electrons in a 110 degree angle and a histogram of all photons emitted from the target was acquired. Different target materials and electron energies were used. The target materials were chosen on basis of the energy of their characteristic fluorescent lines. The energy of the lines should be close to the absorption edge of carbon, yet they should still be able to pass through the primary window. The surrounding medium was set to vacuum for this and all subsequent simulations.

The simulated X-ray tube target spectra were then used as source radiation in a simulation setup illustrated in Fig. 1. In this setup a sample representing cast iron (95.38% Fe, 2.5% C, 1.5% Si, 0.5% Mn, 0.02% S and 0.1% P) was irradiated with a conical beam of photons with a primary window placed between the sample and the source. The primary window was either a 50 μm Be window or a SiN window. The SiN window consisted of a 20 nm SiN layer between two Al layers of 10 nm each and a 15-μm-thick Si grid was attached to it, covering 20% of the window area.

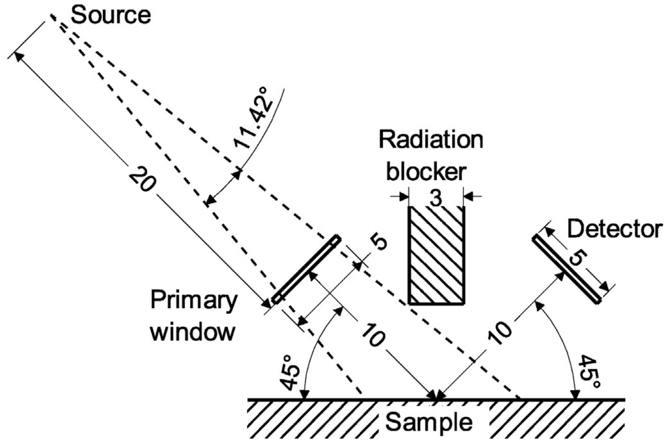

Fig. 1. A sketch of the simulated measurement geometry. The dashed line represents the X-ray source beam. Units are in mm.

Because simulating the entire setup with adequate statistics would take a long time, detector response was simulated separately. In the sample irradiation phase, spectra were acquired for all photons that left the sample in an angle between 30 and 60 degrees with respect to the sample surface. These spectra were then used to generate a photon beam directed perpendicularly into a detector system comprising a silicon slab and a window placed in front of it. The detector window was identical to the SiN primary window described above. The silicon slab was modeled as two layers; a dead layer of 150 nm and an active layer of 500 μm. Spectra for the energy deposition in the active silicon volume were acquired, each incident photon producing at most one recorded event in the detector. In order to take detector resolution into account, the counts were also broadened according to the equation

$$E_{\mathrm{FWHM}} = 2.355\varepsilon\sqrt{\mathrm{ENC}^2 + \frac{FE}{\varepsilon}}$$

where ε is the electron-hole-pair creation energy, ENC the rms fluctuation of the electronic noise, F the Fano factor and E the deposited energy. For silicon, values of $\varepsilon = 3.65$ eV and $F = 0.115$ were assumed. Depending on the composition of the sample and the X-ray tube target material, a very low value of resolution might be needed in order to separate the carbon peak from interfering peaks. State-of-the-art detectors have been reported to have noise values as low as 4.4 e-rms for shaping times that would still allow measurements at high count rates [12]. Therefore ENC was set to 4.4 e-rms, giving a resolution of 46 eV at the energy of C Kα. Since it can be assumed that X-ray tube current could be tuned in order to achieve a desired count rate, the spectra for the energy deposition were normalized to a total of 2,250,000 counts, corresponding to a count rate of 150 kcps and a measurement time of 15 s. The required X-ray tube power to achieve this count rate in the geometry shown in Fig. 1 was also calculated for each combination of an X-ray target and a primary window.

3. Key device comparison

Sensitive X-ray spectroscopy analyses demand the possibility to operate in the wide energy range from soft X-rays to hard X-rays. Screening some part of the range out, typically soft X-rays, limits immediately the sensitivity and reliability of the analyzers. Therefore, a suitable X-ray window needs to be selected. Fig. 2 shows an X-ray transmittance comparison between the most attractive X-ray window choices including polymer, graphenic carbon and SiN based types. These windows have reasonable transmittance over the whole X-ray range and the SiN based window has clearly the highest transmittance over the range from 50 to 2000 eV. The differences among the window types are especially high above carbon K edge of 282 eV where graphenic carbon and polymer windows have lower performance.

Beryllium is the well known workhorse in the X-ray spectroscopy but it has its limitations in the soft X-ray range as can be seen in Fig. 3. Above 5 keV beryllium becomes practically X-ray transparent. However, the same happens to other window types as well but at slightly higher energies. Above 8 keV the difference between 50 μm beryllium and ultra-thin SiN window becomes insignificant. Between 3 and 8 keV beryllium is hard to beat. However, the ultra-thin SiN window has reasonable X-ray transmittance also in this range.

Ultra-thin SiN windows were proven to have excellent radiation hardness against X-rays and they are also known to be leak tight [1–2]. Therefore it is reasonably straightforward to build an X-ray tube with an ultra-thin SiN window. Fig. 4 shows the key benefit of this