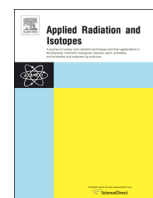




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A new fully integrated X-ray irradiator system for dosimetric research

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HIGHLIGHTS

- Bench top X-ray irradiator provides variable dose-rates.
- Simulation of low energy photon irradiation and hardening of X-ray.
- Al-hardening for the irradiation of H₂O, BeO, Al₂O₃, quartz, feldspars and zircon.
- Dosimetric dating equipment for luminescence and ESR.

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ABSTRACT

A fully housed X-ray irradiator was developed for use within lexsyg or Magnostech desktop equipment. The importance of hardening of the low energy photon radiation is discussed, its performance and feasibility is empirically shown and sustained by basic numerical simulations. Results of the latter for various materials are given for different X-ray source settings in order to provide estimates on the required setup for the irradiation of different geometries and materials. A Si-photodiode provides real-time monitoring of the X-ray-irradiator designed for use in dosimetric dating and other dosimetric application where irradiation of small samples or dosimeters is required.

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1. Introduction

In dosimetric research it is often necessary to calibrate dose-meters or sequentially expose samples to ionising radiation from calibrated sources, e.g., in dosimetric dating application. Depending on the dose, the material or the task, dose-rates differing by several orders of magnitude are desired. The sealed radioactive sources commonly facilitated in commercially available bench top measurement systems, however, can only provide a single fixed dose-rate, or can only be changed to a very limited number of dose-rates, e.g., by use of the bremsstrahlung from closed β -sources or altering the distance to the sample. While higher energy (> 500 keV) photon radiation would be presumably preferred because they are

expected to provide homogeneous irradiation fields, technical reasons, i.e. shielding required for radiation safety as well as the exposure of nearby sample material, limit the use of e.g., γ -sources. Therefore β -sources (e.g., $^{90}\text{Sr}/^{90}\text{Y}$) of limited activity are favoured. But such systems may not provide dose-rates at the desired range for an efficient use of the equipment for different tasks and materials, e.g., personal dosimetry versus medical dosimetry or young versus old samples in dating application.

X-ray tubes are an alternative radiation source because the dose-rates can be varied by altering the tube voltage as well as the tube current. Here, changing the tube current is normally preferable because it does not affect the energy spectrum of the X-rays. This also allows dose-rates that are not easily realized with $^{90}\text{Sr}/^{90}\text{Y}$ -sources, because of a lack of a feasible overall activity in the source design. Also, handling radioactive sources with large activities is usually restricted by legislation and the limited total activity legally permitted in a laboratory. Such restrictions result in low activity sources having to be used for irradiations and thus in

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long irradiation times for large doses. Expensive equipment is therefore inefficiently used for much of its time for the simple procedure of irradiation. If dose-rates can be varied like in X-ray equipment, the exposure times can be shortened by increased dose-rates and thus sample throughput can be increased. Additionally, X-ray sources are favoured from the perspective of radiation safety, because there is no radiation emission when switched off and thus radiation exposure of personnel or samples is minimized, making such equipment less hazardous. Operation of X-ray sources is much easier under many country's legislations, especially as no radioactive source handling is necessary and expensive decommissioning does not apply. Additionally, X-ray irradiators generally provide rather homogeneous irradiations in terms of energy flux over sample area (e.g., Andersen et al., 2003).

The current paper refers to the development of an X-ray source for dosimetric research and application, which is attachable to the *lexsyg* line of luminescence readers from Freiberg Instruments (Richter et al., 2013, 2015), as well as the Electron Spin Resonance (ESR) spectrometer series from Magnettech (e.g., Desmet et al., 2015; Mikou et al., 2015).

2. Constraints of low energy X-ray sources in dosimetric radiation research

While a wide variety of X-ray tubes are employed in many fields of dosimetry, their practical use as part of measurement systems is limited, mainly due to their size and their requirements in power supply, cooling and shielding, making built-in approaches with tubes of several hundred kV not feasible. Therefore the application is limited to X-ray sources of operational dimensions with respect to the size of the measurement equipment (here bench top) and low energy X-ray tubes with voltages up to 50 kV, which were introduced by, e.g., Hashimoto et al. (2002). While the main emission of a $^{90}\text{Sr}/^{90}\text{Y}$ -source are β^- -particles with mean β -energies (Stabin, 2007) of 195.8 keV (^{90}Sr ; $E_{\beta, \text{max}}$: 546 keV) and 933.6 keV (^{90}Y ; $E_{\beta, \text{max}}$: 2280.1 keV; extracted from National Nuclear Data Center, 2015) plus bremsstrahlung with a wide range of photon energies, the most commonly used low energy tubes with 50 kV as maximum voltage can nominally produce X-ray radiation of photon energies of at most 50 keV, with mean energies much below. Dose deposition from a β -source takes place mainly via direct ionisation and is therefore predominantly related to the material density of a sample but almost independent of any other sample properties (e.g., Attix, 1986; Demtröder, 2015). By contrast the dose deposition using low energy X-rays is dominated by the photoelectric effect (cf. Attix, 1986) and is therefore related to the atomic numbers (Z) of the sample compound elements and the thickness of the sample. Its dose deposition is therefore dependent on the composition of the irradiated material. Differences in composition, e.g., the mineralogy of the samples, result in different absorbed doses (e.g., Krieger, 2015). In principle, a calibration for each material and geometry is required. In order to minimize these difficulties, the radiation can be hardened by placing filters (e.g., Al) in the beam. As trade-off, the dose-rate decreases with increasing spectrum hardening. The question of the best compromise between high energy X-ray spectrum and high dose-rate, adjusted by the Al absorber thickness, is thus of crucial importance.

Thomsen et al. (2006a) showed experimentally that the dose-rate dependence on the sample mineralogy and therefore the variance between samples can be reduced by implementation of an Al hardening filter with a thickness of 200 μm . The same optimal thickness is also reported by Yawata (2007), while other values are provided by various authors (50 μm in Hong et al., 2005; 300 μm Kim et al. 2011; Choi et al., 2014), but lacking precise specification of the samples' geometries and compounds to be irradiated.

3. Simulation of low energy X-ray irradiation

In order to provide a better theoretical basis for the development of the X-ray irradiator a simulation of X-ray emissions and absorption was performed using the statistical programming language **R** (R Development Core Team, 2015). It should be noted that it was not intended to precisely model the rather complex interactions physically precise, but rather to provide a framework for application and development allowing the definition of the irradiation set up appropriate for specific geometries, sample materials and sample thicknesses. This also provides a rough guidance on the relative influence of the sample material, its geometry and X-ray source set up parameters.

3.1. Method of simulation

The underlying model is based on an estimated X-ray emission spectrum, consisting of the target-dependent characteristic X-ray-fluorescence emission lines (Krause, 1979; Deslattes et al., 2015) plus a rough estimation of the bremsstrahlung emission spectrum according to Kramer's rule (Krieger, 2015; Laguitton and Parrish, 1977).

The depth of penetration of the tubes electron beam into the anode material is significantly higher than the penetration depth of low energy X-ray photons, resulting in a low escape probability for the low energy tail of the produced X-ray photons. This 'self-attenuation' of the tube was estimated by adding a virtual X-ray absorber made from the same target material with half the thickness of the penetration range of the electron beam. The electron beam's penetration range was estimated according to Katz and Penfold (1952). The emission spectrum is not just altered by the virtual self-attenuation absorber but also by the Be window of the tube, the air between tube and sample and eventually implemented hardening filters. The absorption of all these elements was calculated with the Lambert-Beer-law using the attenuation spectra given by the NIST XCOM: Photon cross section database (Berger et al., 2010). Here the total attenuation values including coherent scattering were used.

The X-ray output was estimated independently by the method given in Compton and Allison (1936) and is used as normalization value for the raw emission spectrum (e.g., the integral of the raw spectrum equals the estimated X-ray output). Based on the assumption that the tube is a Lambert emitter (Brown, 1966), the energy flux through the sample area is calculated. It follows, that the energy flux underlies approximately the $1/r^2$ -distance law and therefore depends on the distance tube to sample and sample diameter.

When calculating the X-ray absorption spectrum, the dose-rate and the penetration depth, the absorption efficiency of the energy flux into the sample is employed according the Lambert-Beer law. The absorption efficiency is determined by the photon cross section spectra from Berger et al. (2010) multiplied with the sample thickness and density. Thus, just photo-ionisation as a dose deposition process is regarded, while Compton-effect, X-ray fluorescence re-emission and scattering effects are ignored. Because the photon cross section and therefore the X-ray absorption efficiency underlies a Z^4 to $Z^{4.5}$ dependency (Z : atomic number; Krieger, 2015), dose-rates and other properties differ significantly between samples of different atomic composition.

3.2. Simulation verification

As no data were available for the Varian VF-50J X-ray tube employed, the published emission spectrum of a similar small X-ray tube (Amptek Mini-X X-ray tube with tungsten target, 40 kV maximum voltage; Amptek, 2015) was compared to its simulated spectrum using the approach described above. The agreement is considered as satisfying (Fig. S1). Furthermore this simulation approach was empirically falsified by spectral measurements with a silica-drift-detector (SDD - RönTec Xflash 1201) of the Varian VF-50J

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