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Characterization of a plastic dosimeter based on organic semiconductor photodiodes and scintillator

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ABSTRACT

Background and purpose: Measurement of dose delivery is essential to guarantee the safety of patients undergoing medical radiation imaging or treatment procedures. This study aimed to evaluate the ability of organic semiconductors, coupled with a plastic scintillator, to measure photon dose in clinically relevant conditions, and establish its radiation hardness. Thereby, proving organic devices are capable of being a water-equivalent, mechanically flexible, real-time dosimeter.

Materials and methods: The shelf-life of an organic photodiode was analyzed to 40 kGy by comparison of the charge-collection-efficiency of a 520 nm light emitting diode. A non-irradiated and pre-irradiated photodiode was coupled to a plastic scintillator and their response to 6 MV photons was investigated. The dose linearity, dose-per-pulse dependence and energy dependence was characterized. Finally, the percentage depth dose (PDD) between 0.5 and 20 cm was compared with ionization chamber measurements.

Results: Sensitivity to 6 MV photons was (190 ± 0.28) pC/cGy and (170 ± 0.11) pC/cGy for the non-irradiated and pre-irradiated photodiode biased at −2 V. The response was independent of the dose-per-pulse between 0.031 and 0.34 mGy/pulse. An energy dependence was found for low keV energies, explained by the energy dependence of the scintillator which plateaued between 70 keV and 1.2 MeV. The PDD was within ± 3% of the ionization chamber.

Conclusion: Coupling an organic photodiode with a plastic scintillator provided reliable measurement of a range of photon energies. Dose-per-pulse and energy independence advocate their use as a dosimeter, specifically image-guided treatment without beam-quality correction factors. Degradation effects of organic semiconducting materials deteriorate sensor response but can be stabilized.

1. Introduction

Development of novel materials and portable devices are essential to determine radiation dose accurately in many applications including radiation safety and medical diagnostics. High spatial resolution for precision, instant feedback for radiation safety and mechanical flexibility for use with complex contoured surfaces are challenges that have not been solved by the current technology used. Organic semiconductors are a particularly attractive class of materials for advanced dosimetry purposes, as they can combine the electronic advantages of semiconducting substrates with the chemical and mechanical benefits of organic compounds. As these materials are composed almost entirely of carbon, hydrogen, and oxygen, they have a response to radiation that closely mimics that of water. Furthermore, organic materials can be dissolved in solutions to create inks. This introduces avenues for printing the electronic materials directly onto mechanically flexible substrates at high speeds across large areas [1,2]. This printable manufacturing pathway also allows organic semiconductors to provide flexible and wearable low-cost devices [3]. Although the use of organic electronic materials for applications as Light Emitting Diode (LED) displays [4,5] and photovoltaic devices [6] are mature research fields, very little is presently understood about their response to exposure to

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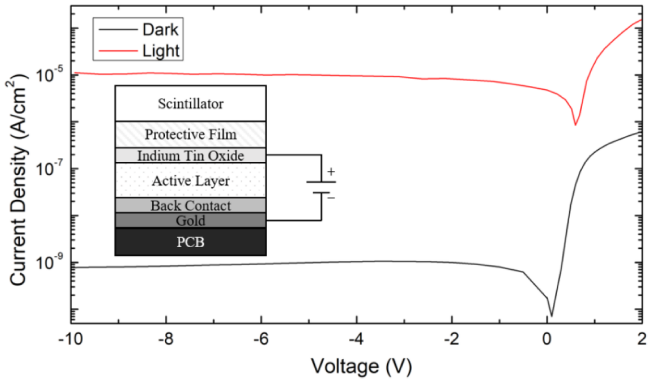

Fig. 1. Electrical characteristics of the photodiode in dark conditions and illuminated at 530 nm. An inset illustrates the structure of the photodiode and bias configuration.

high energy ionizing radiation. Fully organic semiconductors have shown promising applications as X-ray imagers via the exposure to 70 keV X-rays [7,8] and as dosimeters for 6 MV photon fields [9,10]. These works have also observed minimal degradation in performance up to 500 Gy. However, further work is required to ensure they can accurately measure X-rays in a variety of clinically relevant characterization techniques and have a long shelf life with high exposure to ionizing radiation fields. Thus, the purpose of this work is to investigate the usability of an organic scintillator, readout by an organic photodiode, for clinical applications. This work also observed the variation in charge collection efficiency when exposed up to an accumulated dose of 40 kGy to investigate the effects of radiation damage on their ability to accurately measure ionizing radiation.

2. Materials and methods

2.1. Detector structure and Electrical characterization

An Organic Photodiode (OPD) was produced by ISORG (Grenoble – France) and features a 500 nm thick bulk heterojunction with an active area of 4.91 mm^2. The basic structure, illustrated from the inset in Fig. 1, consists of an indium tin oxide transparent anode deposited on the top side of the substrate allowing light to penetrate the active layer and a back contact attached to a gold pad. The photodiode was covered with a protective film (Polyethylene, 25 μm) to minimize material oxidation and mounted onto a printed circuit board (FR-4, 2 mm) for ease of connection to the readout electronics. The current density-voltage characteristics were measured using a Picoammeter/ Voltage Source (Keithley 487) applying bias from −10 V to 2 V, in dark conditions and illuminated by a 530 nm LED.

2.2. Radiation damage

To characterize the damage of the OPD, the response to visible light was characterized as a function of accumulated radiation exposure. The OPD was irradiated by a cobalt-60 gamma source (dose rate = 1.2 l kGy/h). During irradiation, the OPD was embedded in a plexiglass holder with a 1.5 cm thick window between the gamma source and sample surface for face-on (detector surface perpendicular to beam) irradiation. Irradiation occurred in 5 steps from 0.1 kGy to 1 kGy in steps of 0.25 kGy with further larger steps of 10 kGy to reach a total ionizing dose of 40 kGy. The remaining non-irradiated OPDs were used as reference samples. The diodes adopted in this study are part of a commercial production batch for an imaging device. The industrial process guarantees high quality and response reproducibility between samples with leakage currents within ± 5% of the total bias range, allowing the non and pre-irradiated diodes to be directly compared. At each step the charge-collection-efficiency of the photodiode was measured using an LED at 520 nm in a light tight box. The LED was mechanically chopped at 190 Hz and attenuated via a neutral density filter. Signal averaging over multiple pulses and baseline subtraction was carried out to improve the measurement accuracy and evaluate uncertainties. Radiation damage studies were conducted in response to light because further measurements utilize the detection of X-rays in combination with a plastic scintillator.

2.3. Dosimetry characterization

Photon dose response measurements were performed using 6 MV X-rays from a Varian Clinac 21iX Linear Accelerator. The Linear Accelerator is a pulsed radiation source with pulse length of approximately 3.6 μs, repetition rate of 360 Hz and machine output rate of 600 MU/min. Detection was achieved by coupling the photodiode with a plastic scintillator (Rexon, RP400) converting X-rays and secondary Compton electrons into visible light with a peak emission wavelength of 420 nm with a broad spectrum up to 600 nm to match the sensitivity of the OPD to the visible spectrum. The scintillators thickness was varied between 0.5 mm and 2 mm depending on the measurement conducted and were coupled to the photodiode with optical grease (Rexon, RX-688). The thickness of the scintillator plays an important role to minimize the effect of the current generated by direct interaction of the secondary electrons and primary X-ray photons with the diode's substrate. For each measurement the devices were embedded within a 30x30x1 cm^3 plexiglass holder for irradiated face-on and connected to a custom designed electrometer for real-time measurements [11]. An external reverse bias of −2 V was applied at the photodiode. Reference conditions for the 6 MV photon beam were a source to surface distance (SSD) of 100 cm, field size of 10x10 cm^2 and sample depth of 1.5 cm in a water equivalent plastic phantom (1 MU = 1 cGy), with an additional 10 cm backscatter. Dose linearity measurements were performed at reference conditions by depositing 25 cGy to 500 cGy of 6 MV photons at reference conditions for both the pre and non-irradiated devices coupled with a 0.5 mm thick scintillator. A black cloth was placed over the phantom and the baseline was subtracted to account for any ambient light. Each measurement was repeated three times to obtain an average and standard deviation. Detector sensitivity was calculated from the gradient of the linearity plot (charge vs accumulated dose). The collected charge when coupled with a 2 mm thick scintillator at 1.5 cm depth in the phantom, relative to an ionization chamber (NE2571 Farmer) in the same conditions, was tested under different irradiation dose rates by varying the SSD from 300 cm to 90 cm, measured with a precise laser rangefinder, for 100 MU of 6 MV photons. The intensity of the photons decreases as one over the square of the distance, changing the intensity of the flux of photons reaching the detector but with negligible effects on the spectrum of the beam. This corresponds to a dose per pulse range between 0.031 and 0.34 mGy/pulse, respectively. The ratios were normalized to the collected charge ratio measured at an SSD of 100 cm (0.28 mGy/pulse). For energy dependence measurements, the collected charge of the photodiode coupled with a 2 mm plastic scintillator to a dose of 100 cGy was compared for 6 MV photons in reference conditions and keV X-ray energies produced from a calibrated Gulmay orthovoltage X-ray tube. The accelerating potentials of the X-ray tube were varied between 50 kVp and 100 kVp corresponding to incident average photon energies of 29.5 keV and 50.0 keV using an aluminum filter. For higher energies, a copper filter was used for accelerating potentials of 150 kVp to 250 kVp to achieve incident photons energies of 79.5 keV to 129.4 keV. X-ray tube measurements deposited 100 cGy onto the photodiodes at 30 and 50 cm focus to skin distances, depending on the applicator used for the desired energy, at surface with 10 cm backscatter of solid water. The ability of the detector, when coupled with a 1 mm scintillator, to reconstruct the dependence of the dose deposited from a 6 MV photon beam as a function of depth was measured. The depth within the

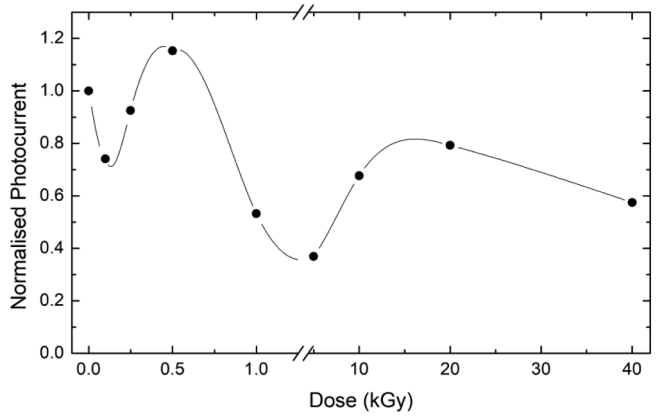

Fig.2. Charge collection efficiency of an irradiated organic photodiode from a pulsed 520 nm LED, normalized to the response before irradiation. No scintillator is attached to the photodiode in this study. Uncertainties (one standard deviation) are smaller than the symbols.

phantom was varied from 0.5 cm to 20 cm with a constant SSD of 100 cm and normalized to the reference conditions, i.e. 1.5 cm depth, to compare with the percentage depth dose (PDD) of a Scanditronix/Wellhofer CC13 ionization chamber.

3. Results

Full charge extraction was observed from a reverse bias of −2 V, as shown in Fig. 1 and was therefore used as the bias voltage for the response measurements. Under illumination the photocurrent was stable at $(1 \pm 0.3) \times 10^{-5}$ A/cm^2 from 0 V to −10 V.

Radiation damage studies of the photodiode were investigated up to 40 kGy. Fig. 2 shows a priming effect during the first 100 Gy with an increase of 15% in response followed by significant degradation up to 5 kGy. After 10 kGy the response stabilized at a charge collection efficiency of 65%, remaining within ± 10% up to 40 kGy.

The dose linearity for the photodiode coupled with a 0.5 mm scintillator was achieved prior to and following exposure to 40 kGy (Fig. 3). The non-irradiated device was determined to be (190 ± 0.28) pC/cGy with an R^2 value of 0.9991. Pre-irradiation resulted in a 12% decrease of the sensitivity with a value of (170 ± 0.11) pC/cGy and R^2 value of 0.9999. The normalized collected charge, presented in Fig. 4, showed a uniform dependence on dose per pulse within a relative standard deviation of 6% of the total range tested. There was also a negligible effect on dose per pulse dependence from radiation damage, suggesting that

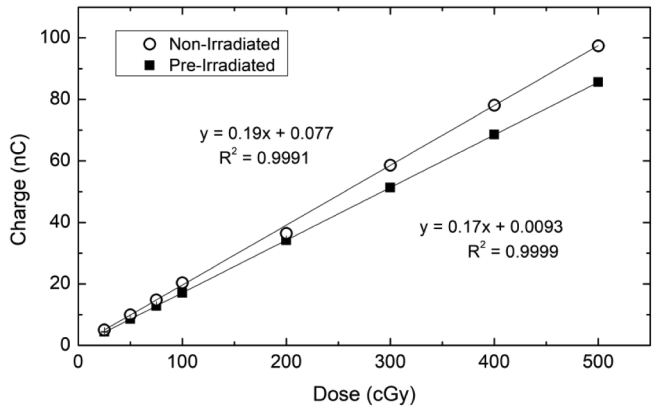

Fig. 3. Charge response to 6 MV photons from an organic photodiode, non-irradiated and pre-irradiated to 40 kGy of ionizing radiation, coupled with a 0.5 mm thick plastic scintillator. Uncertainties (one standard deviation) are smaller than the symbols.

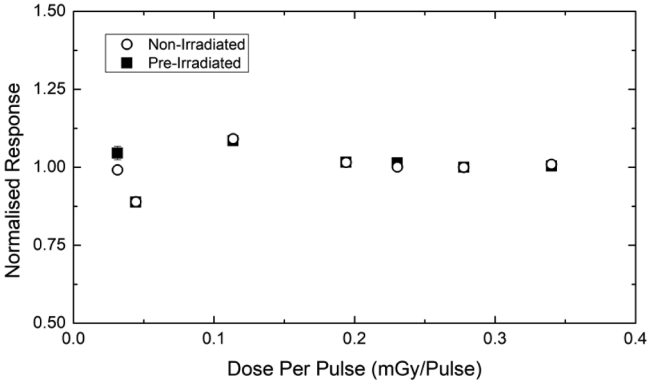

Fig. 4. Uniform response to the variation of the dose per pulse of 6 MV photons with non-irradiated and pre-irradiated photodiode coupled with a 2 mm thick plastic scintillator. Uncertainties (one standard deviation) are smaller than the symbols.

the ratio between diffusion and drift of carriers is not disturbed by the presence of radiation induced defects in the substrate. Uniform dose per pulse dependence allowed for the energy dependence measurements to be achieved as the dose rate of the X-ray tube varied with the desired energy output between 9 mGy/s to 42 mGy/s. The only effect of the dose rate was the time required to deposit 100 MU, which was deposited for each measurement, and can be correlated with the larger error bars obtained at 100 keV which possessed the lowest dose rate. The energy dependence of the detector coupled with a 2 mm thick scintillator was obtained for a range of keV energies and compared to the charge collected from 6 MV photons (Fig. 5). The gradient from 30 keV to 100 keV is due to the absorption of the plastic scintillator as proved by comparison with the scintillator's material absorption within the measured energy range. Fig. 6 presents the PDD for the photodiode coupled with a 1 mm thick scintillator between a depth of 0.5 cm to 20 cm in solid water. The relative difference for the non-irradiated photodiode was within ± 3% of the ionization chamber.

4. Discussion

In this study we have shown the potential of using organic photodiodes combined with a plastic scintillator as radiation detectors for radiotherapy treatment applications. In order to assess its usability for MV beams we measured the collected charge to variations in depth in solid water, and variations of dose rates. The radiation hardness of the organic semiconducting material was also characterized for an accumulating dose orders of magnitudes larger than any previous

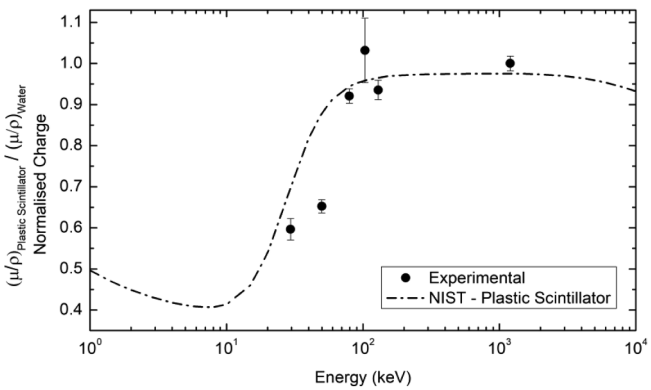

Fig. 5. Response of an organic photodiode coupled with a 2 mm thick scintillator to keV X-rays, normalized to the response of 6 MV photons with an ⟨E⟩ = 1.2 MeV. In comparison to the response of the plastic scintillator via NIST [22]. Uncertainties were defined as one standard deviation.

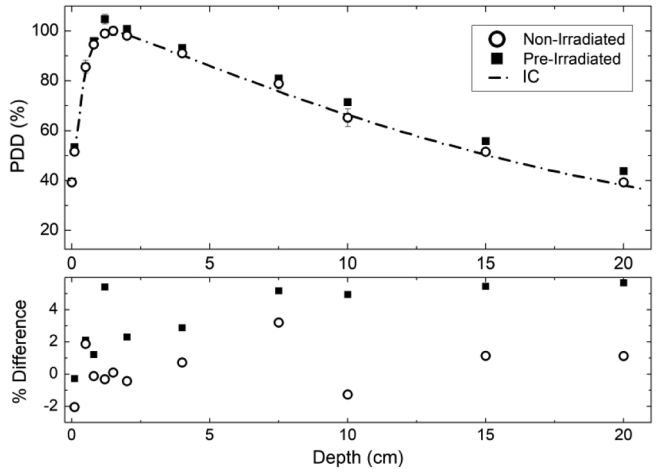

Fig. 6. PDD of the organic photodiode, non-irradiated and pre-irradiated up to 40 kGy, coupled with a 1 mm thick plastic scintillator. The percentage difference to an ionization chamber is compared. Uncertainties were defined as one standard deviation.

investigations.

Organic semiconductors are outlined to be highly advantageous in comparison to inorganic dosimeters. This is due to their ability to be printed into large areas via low-cost methods onto flexible substrates, thus giving them the ability to conform to the human body for in-vivo dose verification and 2D dose mapping. Since organic materials are composed of mostly carbon and hydrogen, they interact with ionizing radiation in a similar fashion to the human body. The density of the scintillator used was 1.023 g/cm^3 with an electron density of 3.37x10^{23} cm^{-3} compared to 3.34x10^{23} cm^{-3} for water [12]. This is one of the motivations behind using plastic scintillators and have consequently been extensively studied in terms of their radiation hardness and dosimetry applications [13,14]. Therefore, if the scintillator is coupled with an organic photodiode, the entire device can be fabricated from materials that are water equivalent. Otherwise, the presence of high-Z materials around the sensitive volume perturbs the energy fluence of the particles. Thus, the readout system that is coupled directly to the scintillator must also be removed of high-Z materials, otherwise this will generate a dose enhancement via scattering at the interface, affecting the response of the scintillator.

In order to establish if the combination of plastic scintillator and organic photodiode can be used for dosimetry, we investigated the organic semiconductors variation in charge collection efficiency due to the exposure to ionizing radiation i.e. its radiation hardness. As expected, radiation damage affects the transport of the carriers in the substrate, leading to a decrease of the responsiveness of the device. The response stabilized at approximately 65% of the initial sensitivity, with a fluctuation of ± 10% due to annealing effects. The annealing represents an important parameter and contributed to the effect of relaxation of the material after exposure to intense radiation. The annealing effects are greatly affected by the dose rate of the irradiation field. The dose rate used for this investigation falls into a class of high dose rates that produce high distribution of atoms in temporary excited state in the semiconducting material, resulting in drastic annealing effects [15]. The effects become stable with time allowing the material to recover. Self-recovery in organic molecules has been shown in other literature to improve several hours after irradiation of 100 Gy. The time delay between irradiating the samples and measuring the devices collected charge increased for larger step sizes past 5 kGy. This limitation was due to the need to leave the devices in the irradiator overnight. A consistent timing between irradiation and testing needs to be taken into consideration for future measurements to minimize the effect of annealing on the interpretation of the results. While the uncertainty in their charge collection efficiency is 10% past 10 kGy, previous literature reported that organic semiconductors fail to respond at a high total irradiation doses. Therefore, this result demonstrates that organic semiconductors can perform in these harsh environments with high radiation tolerance. It is also worth noting that these devices have been tested for over four years, exposed to a range of electrons, gamma and X-ray beams, and stored in a dark, air permeable environment; indicating a potential long shelf-life if used as a dosimeter.

The photodiodes ability to readout the visible light generated from a scintillator in clinical conditions was then investigated as a preliminary study to propose their potential as a dosimeter. The sensitivity of the photodiode coupled with a 0.5 mm thick scintillator was observed to decrease by 12% after pre-irradiation, while the linearity dependency was not affected (Fig. 3). The combination of the plastic scintillator and the photodiode achieved a dose per pulse dependence that are within a relative standard deviation of 6% across the tested range. This is a remarkable result in respect to other technologies such as diamond [16,17] or silicon diodes [18] already used clinically. The dose per pulse with the greatest deviation was between 0.03 mGy/Pulse to 0.11 mGy/Pulse, reaching up to −12%. These values correlate to widely out of field dose deposition at a very large depth in a water equivalent phantom limiting the effect on the accuracy obtainable with the detector. The dose per pulses within the range of 0.19 mGy/pulse to 0.34 mGy/pulse (standard conditions at 6 Gy/min and repetition rate of 360 Hz leads to 0.27 mGy/pulse) are within 0.79% variation for the non and pre-irradiated devices. These results suggest that while the preliminary results are promising, the dose per pulse dependence may require some corrections for clinical use and will need to be investigated for higher dose rates from a Flattening Filter Free Linear Accelerator.

The energy independence of the fluorescence of a plastic scintillator readout by an organic photodiode has also been quantified for keV and MeV energy range photons (Fig. 5). The energy dependence from this X-ray source combines the effect of the monoenergetic beam used, and the influence of effects such as quenching of the scintillator response to low energy electrons [19–21]. While the theoretical data obtained from NIST for a RP-400 scintillating material does not include these effects, its comparison with the experimental data shows that the relationship of the energy dependence is dominated by the energy dependence of the attenuation in the plastic scintillator, inferring energy independence of the organic semiconducting material. After 100 keV the energy dependence plateaus, demonstrating the advantage of using organic detectors for dosimetry techniques that require varying X-ray energies e.g. imaging guided radiotherapy.

The PDD curve was shown to be affected by the direct interaction of the photodiode, producing an overresponse in Fig. 6 due to its position directly under the beam. The contribution of the total signal from the direct Compton electron interaction was close to negligible when coupled with a 1 mm thick scintillator, but when proposed as a dosimeter it may need to be corrected for. The PDD measured by the heavily pre-irradiated photodiode in respect to the IC showed a discrepancy within ± 6%. This was not observed for the non-irradiated device which showed, in contrast, a discrepancy less than ± 3%.

The results suggest that the inerrant damage of the material, due to the total irradiation dose of 40 kGy, has impacted the ability of the device to accurately measure the dose deposited in direct comparison to an IC. An optimum pre-irradiation dose needs to be identified to produce a stable device that does not suffer from degradation, and still have a sufficient long shelf-life and response stability. This pre-irradiation value was observed to be identifiable at 10 kGy when a charge collection degradation of 68% was achieved (with a final degradation of 65% expected at 40 kGy). An extension of this characterization to direct detection and other radiation dosimetry parameters, such as output factor and irradiation by electrons, will also be valuable. An understanding of the effects that govern the radiation damage in organic semiconductors is also required identifying the design parameters to

optimize these devices for dosimetry.

In conclusion, this study characterized the response of an organic photodiode coupled with a plastic scintillator to both keV and MeV X-rays for use as a dosimeter and effects of radiation damage. Device sensitivities reached (190.0 ± 0.3) pC/cGy with a uniform charge collection to dose per pulse dependence and an energy dependence influenced only by the scintillator. Radiation damage studies proved the devices possess a long shelf-life, after a stabilizing pre-irradiation step. The findings suggest that these devices can be used for real-time measurements with minimal perturbation of the beam, demonstrating to be potential candidate as a water equivalent, flexible substrate for dosimetry.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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