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Gamma-ray response function of some polymeric nuclear track detectors

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The effect of gamma dose on the bulk-etch rate measurements for some gamma-irradiated polymeric nuclear track detectors was extensively studied. Various plastic samples of CR-39 allyl diglycol carbonate, Makrofol-E and Lexan polycarbonates were used and exposed to gamma doses up to 30.0 Mrad. The bulk-etch rate ratio V_{BD}/V_{BO}, i.e. the bulk-etch rate of irradiated samples to the unirradiated ones, was measured at the most recommended optimum etching conditions. Fading behaviour of irradiated CR-39 samples was also studied at room temperature and for a duration of up to 8 days. It was found that the etch-rate ratio for the CR-39 specimens was dose-dependent. In contrast, the polycarbonate samples showed an extremely weak response to gamma irradiation. The results of the present work reflected great evidence of the gamma dosimetric potential of CR-39 plastic detectors, which can indeed be recommended as gamma-dosimeters within the studied dose range.

1. Introduction

Radiation effects in solids are a subject of great importance and sometimes also an emotional one. Different techniques [1–6] have been developed for radiation measurement in solids. Moreover, the use of solid-state nuclear track detectors in the identification of charged particles is one of the most significant and amazing techniques in radiation dosimetry [5, 7–10]. The regions of applicability of nuclear track detectors have grown rapidly; they have been used in various fields [11–20], such as charge and energy information of light and heavy nuclear particles, cosmic ions charge discrimination, neutron personal monitoring systems, uranium and radon determination in rocks, water and plants, etc.

The interaction of high-energy radiation, such as gamma rays, with solid material depends on the nature of such an absorber; but the main effect is to produce ionization and excitation of atoms or molecules. Some solid materials, such as glasses, change colour [21] when exposed to gamma radiation and, as a result, colour centres would be produced which lead to a change in the internal electronic configuration of the material. Such centres caused absorption at particular wavelengths and have been detected through optical density measurements. The glassy materials have been successfully used in the field of radiation dosimetry, where several authors [21–24] have tested different glass compositions as gamma dosimeters. More recently [25–28], the effect of gamma rays on some phosphate glasses have been inclusively studied by one of us, and the results have proved the necessity of using the examined glass systems as gamma-ray dosimeters in the studied domain.

The main effect of gamma interaction with plastic detectors is attributed to the degradation of the material which leads to the reduction of the average molecular weight of the detector. Such reduction in molecular weight would be expected to show an increase in the bulk-etch rate, V_B, as the gamma dose increases. V_B is simply defined as the velocity with which the nuclear track detector is dissolved during the period of etching. More details will be forthcoming in Section 3.

Recently, the use of some solid-state nuclear track detectors as gamma-ray dosimeters has been tested [29, 30], but the data available for all kinds of plastics are not quite sufficient. For example, Zamani *et al.* [30] have studied the behaviour of some track detectors to gamma radiation at a dose rate of about 10 krad min^{-1}. They proved that the CR-39 foils (Pershore Ltd, UK) were the best gamma-ray dosimeters of all the other studied materials. In addition, the effect of gamma dose rate on the CR-39 plastic detector has been investigated and reported previously [31]. It was found that the induced changes of etch rate ratio (V_{BD}/V_{BO}) was not pronounced for dose rate ranges from about 3.0–10^2 krad min^{-1} and for a total exposure dose up to 800 krad.

The response of a detector to nuclear radiation is basically dependent on the interaction mechanism between the incident radiation and the internal structure of such a detector. Also, the sensitivity of a detector to any kind of radiation is essentially a strong function of the measured radiation-induced effect in the detector. If the measurable induced effect is sufficiently effective, the sensitivity of the detector would be reasonably high.

In this investigation, an extensive study of gamma

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dose effect on some selected polymeric materials was carried out in order to measure the induced effects and to investigate the possibility of using such plastics as gamma-ray dosimeters. Fading properties of these gamma-irradiated plastic detectors were also detected and studied as a function of storage periods at room temperature and 135 °C.

2. Experimental procedure

2.1. Samples

Plastic sheets of CR-39 allyl diglycol carbonate 1.5 mm each thick (Homalite, Wilmington, DE, USA) were used. Sheets of Makrofol-E (Bayer AG, Germany) and Lexan polycarbonate (General Electric Co., USA) of thicknesses 300 and 500 μm, respectively were also used. All plastic sheets were cut into pieces of about 6 cm^2 in area and exposed to gamma radiation of various doses up to 30 Mrad.

2.2. The bulk-etch rate, V_B, measurements

In general, etching is a diffusion process and the etch rate is essentially dependent on the type, temperature, concentration of etchant solution and the duration of the etching process. The measurement of bulk-etch rate, V_B, of a plastic foil is very necessary for studying the material properties. In fact, there are various methods [10] for V_B measurements. Throughout the present work, a reliable and convenient method was used, based on measuring the mass change of the plastic material as a result of etching. In this technique, each piece of material was carefully weighed using an analytical balance of an accuracy of 0.1 mg before and after etching. Then the sample's mass decrement, Δm, dissolved in the etchant solution, during a period of time, t_e, was evaluated. The bulk-etch rate was calculated from the relation

$$V_B = \Delta m / 2 A \rho t_e \quad (1)$$

where A is the surface area of the detector (cm^2) and ρ is its density ($g\,cm^{-3}$).

In the case of CR-39 plastics, where a sample thickness of about 1.5 mm was used, a side area correction factor was included in the above relationship. For bulk-etch rate determination a Julabo water bath, of temperature accuracy ± 0.5 °C, was used. After etching, samples were rinsed in running water for more than 5 min in order to remove all etch products from the surfaces and then thoroughly washed with distilled water for a sufficient period using an ultrasonic cleaner. Finally, samples were carefully dried using a cold-air dryer before they were weighed again for weight decrement evaluation.

2.3. Irradiation facilities

The plastic sheets used in this study, were exposed in air to different gamma-doses using a ^{60}Co gamma cell as a source of gamma radiation. The exposure rate of 10.35 $krad\,min^{-1}$ was applied at room temperature.

3. Results and discussion

The bulk response of a track detector to gamma radiation was determined via the relative bulk etch rate, V_{BD}/V_{B0}, measurements, where V_{BD}/V_{B0}, is defined as the bulk etch rate of irradiated specimens to the unirradiated ones. Such a method is very reliable and certainly useful to minimize, to a large extent, the experimental uncertainties that would be expected in the absolute bulk-etch rate measurements. The irradiated and unirradiated samples were etched in the same etchant solution at the same time. The variation of the bulk-etch rate ratio with gamma-ray doses was studied under different etching concentrations and at a temperature of 70 °C.

Samples of CR-39, Lexan and Makrofol-E polycarbonate were exposed to various gamma doses up to 30 Mrad. For CR-39 sheets and for gamma-doses ranging up to 2.5 Mrad, the bulk-etch rate ratio was measured as a function of gamma dose. Within such a range of gamma doses (0–2.5 Mrad) the bulk response of CR-39 detector, as measured by V_{BD}/V_{B0} ratio was not linear, which reflected the difficulty of using such a material as gamma dosimeter in the 0–2.5 Mrad dose range. Samples were etched in 5.25 N NaOH at 70 °C. Moreover, V_{BD}/V_{B0} measurements were also performed at various normalities of NaOH solution, for samples irradiated up to 2.5 Mrad, and the results also showed no linearity behaviour of the V_{BD}/V_{B0} ratio with gamma dose.

Fig. 1 shows the dependence of relative bulk etch rate on gamma-dose values begins at 3.0 Mrad and extends up to 20 Mrad for CR-39 Homalite plastic samples etched in 5.5 N and 6.25 N NaOH solutions at an etch temperature of 70 °C. From inspection of

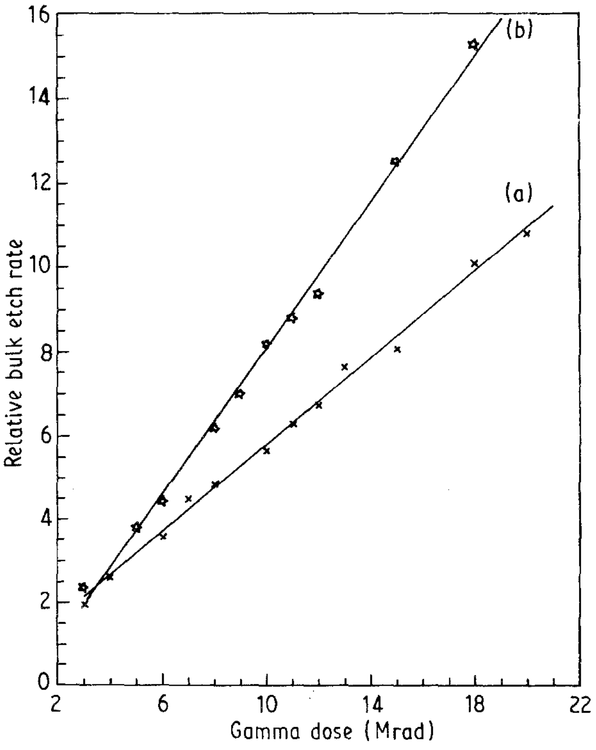

Figure 1 Variation of relative bulk etch rate V_{BD}/V_{B0} with gamma dose for CR-39; samples etched in (a) 5.5 N and (b) 6.25 N NaOH, at 70 °C.

Fig. 1, it is clear that the V_{BD}/V_{BO} ratio is basically a strong function with gamma dose; it shows an increase with increasing gamma dose. Also, the linearity behaviour resulting from the data of Fig. 1 is greatly important in radiation dosimetry studies.

Fig. 2 shows the results of V_{BD}/V_{BO} measurements with gamma dose using CR-39 detectors etched in 7.0 N and 8.0 N NaOH at 70 °C, while in Fig. 3 the V_{BD}/V_{BO} measurements were determined using etchant normalities of 9 N and 10 N NaOH at the same etch temperature of 70 °C. Regarding the results presented in Figs 1–3, it is very clear that there are obvious detectable changes of the V_{BD}/V_{BO} measurements as a function of gamma dose. It is also interesting to note that the linearity behaviour is extended up to a gamma dose value of 20.0 Mrad in case of CR-39 samples etched in 5.5 N and 6.25N NaOH (see Fig. 1). However, as shown in Fig. 2, the situation is not the same where the dependence of V_{BD}/V_{BO} on the gamma dose is only linear up to 10.0 Mrad. Moreover, the linearity dependence given in Fig. 3 consists of two segments, from 3.0–7.0 Mrad and from 7.0–10 Mrad, as in the case of CR-39 samples etched in 9 N NaOH solution. In Fig. 3b (samples etched in 10 N NaOH), the variation of V_{BD}/V_{BO} with gamma dose values from 7.0–10.0 Mrad was found to be an increasing power function rather than a linear relationship. It is obvious that the induced changes of V_{BD}/V_{BO} from 7–10 Mrad (Fig. 3) are more pronounced than those measurements in dose range from 3.0–7.0 Mrad.

In general, results of the V_{BD}/V_{BO} measurements were fitted to an empirical linear relationship given by

$$V_{BD}/V_{BO} = a_n D + b_n \qquad (2)$$

with a correlation coefficient better than 99.8 %, where D is the absorbed gamma dose in Mrad, a_n and b_n are the fitting constants. The subscript n stands for the normality of the etchant solution. Values of a_n and b_n are given in Table I.

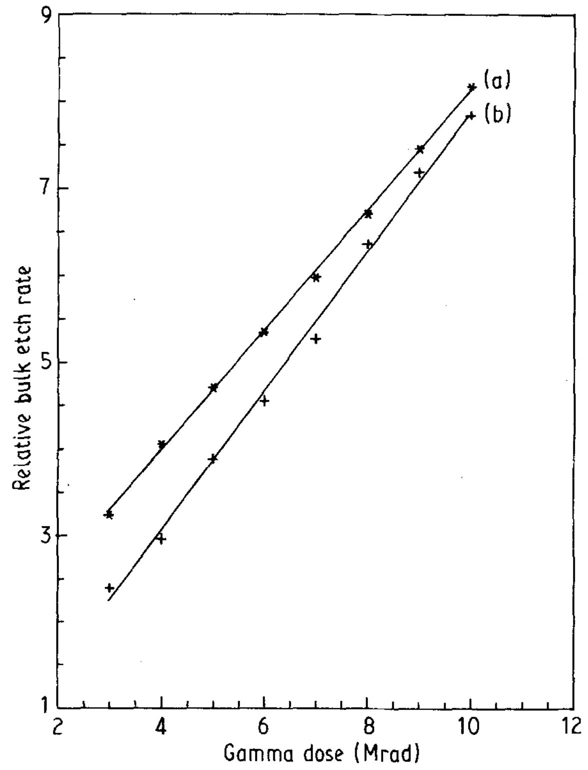

Figure 2 Variation of relative bulk etch rate V_{BD}/V_{BO} with gamma dose for CR-39; samples etched in (a) 7 N and (b) 8 N NaOH, at 70 °C.

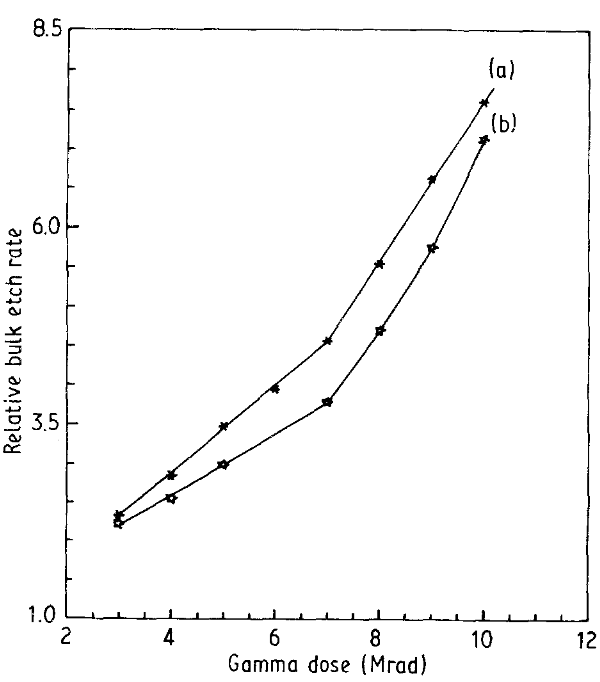

Figure 3 Variation of relative bulk etch rate with gamma dose for CR-39; samples etched in (a) 9 N and (b) 10 N NaOH, at 70 °C.

TABLE I Values of the fitting constants (a_n and b_n) included in V_{BD}/V_{BO} measured as a function of NaOH normality

N	a_n	b_n	Dose range (Mrad)
5.5	0.5185	0.5634	3–20
6.25	0.8666	– 0.6304	3–20
7	0.6949	1.1895	3–10
8	0.8043	– 0.1711	3–10
9	0.5539	0.6659	3–7
9	1.0179	– 2.5729	3–10
10	0.3919	1.0179	3–7

From all previous measurements of V_{BD}/V_{BO} ratio, it was clear that the induced changes in the V_{BD}/V_{BO} measurements were dose dependent; they show a significant increase with increasing gamma-dose (see Figs 1–3). This behaviour can be explained from the point of view of gamma interaction with the internal structure of the polymeric samples. Such an interaction leads to ionization and excitation processes of atoms or molecules that form the detector material, via the secondary products of the interaction. Because the constituents of the polymeric compound are of low Z-value (hydrogen–oxygen and carbon), then the most dominant interaction of gamma rays with such a material is of the Compton scattering type [5]. In such a reaction, ejection of secondary electrons from the absorbed materials would occur and many ionized and excited atoms or molecules would result. The amount of ionization or excitation would produce the detectable effects in the material foils and could serve as a measure of the dose-value absorbed by such materials. As a result, numerous broken molecular chains and free radicals will be produced which leads to a reduction in the molecular weight of the absorber materials [7]. Therefore, the etchant solution will accelerate and easily diffuse through the materials and this may provide an explanation for the behaviour (an increase) of the V_{BD}/V_{BO} ratio as a function of absorbed gamma dose.

Fortunately, the linearity dependence shown in Figs 1–3 tends to indicate the possibility of using the CR-39 samples as gamma ray dosimeters. Such a material really shows an excellent detection ability, especially in the dose range 3.0–20.0 Mrad.

For comparison, some other track detectors have been tested as gamma-ray meters. Fig. 4a and b show plots of V_{BD}/V_{BO} measurements against gamma dose up to 30.0 Mrad for Makrofol-E and Lexan polycarbonate foils, respectively. The Makrofol-E and Lexan detectors were etched using the most recommended etching conditions of PEW etchant reagent [10] (15 g KOH + 40 g water + 45 g ethyl alcohol) at 70 °C. It is clear from Fig. 4 that even the V_{BD}/V_{BO} ratio is still more than one, but the dependence of this ratio on the gamma dose value is unfortunately complex. The V_{BD}/V_{BO} value shows an increase as one goes from 0.0–0.5 Mrad (see Fig. 4) and then falls with further increase of gamma dose up to 2.5 Mrad. Beyond this value of gamma dose, the V_{BD}/V_{BO} value slowly increases, but not in a linear fashion, as gamma dose increases up to 30 Mrad. In addition, the overall rate of increase in the V_{BD}/V_{BO} value is much smaller than that observed in the CR-39 foils. For example, and as shown in Fig. 4b, only a change of about 7.0 % in V_{BD}/V_{BO} value resulted as the gamma dose value increased from 10.0 to 18.0 Mrad. Comparing with CR-39 foil (see Fig 1a), a detectable change of approximately 47.0 % in V_{BD}/V_{BO} value was obtained during the same interval of gamma dose.

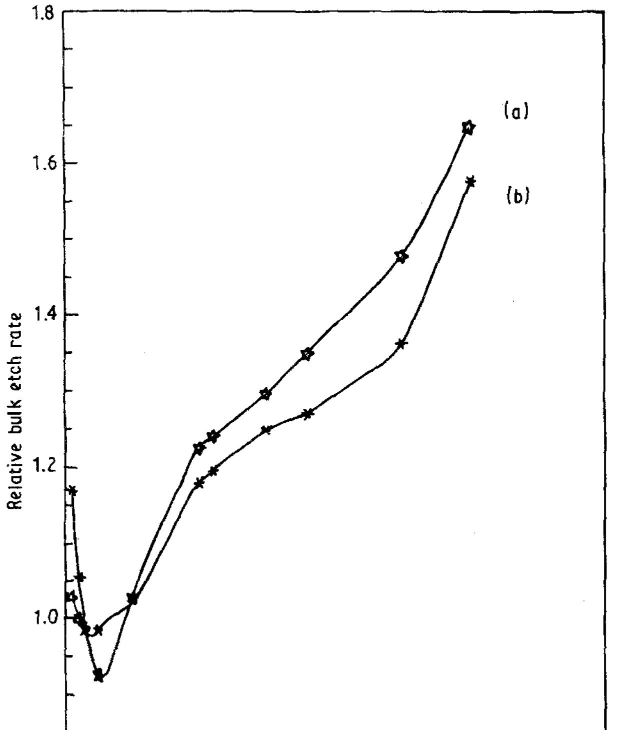

Figure 4 Dependence of relative bulk etch rate V_{BD}/V_{BO} on gamma dose for (a) Makrofol E and (b) Lexan etched in PEW solution at 70 °C.

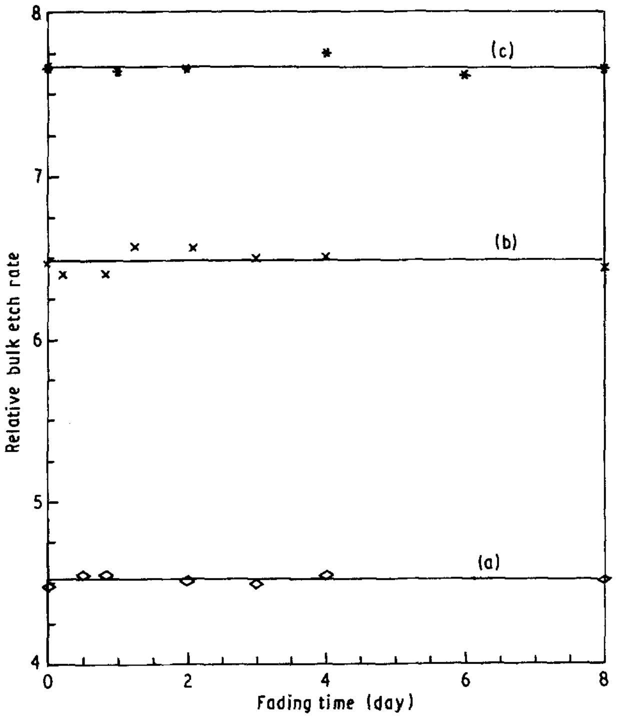

Figure 5 Effect of fading time at room temperature on the relative bulk etch rate V_{BD}/V_{BO} measurements of CR-39 detectors, etched in 5.5 N NaOH; samples preirradiated at (a) 7, (b) 11 and (c) 13 Mrad.

From inspection of Fig. 4, it can be seen that not only are the induced changes in the V_{BD}/V_{BO} ratio a remarkably weak dose-dependent function, but also the linearity dependence is lost. Such results caused obvious difficulties in using either Makrofol-E or Lexan plastics as gamma dosimeters.

A study of the fading behaviour of CR-39 plastic foils is very important for the practical applications of such material in the gamma dose measurements. In this work, fading was studied at room temperature and 135 °C for various times. Fig. 5 illustrates a typical fading characteristic, where the V_{BD}/V_{BO} ratio is plotted against storage time up to 8 days at room temperature using CR-39 samples exposed to gamma doses of 7.0, 11.0 and 13.0 Mrad and etched in 5.5 N NaOH at 70 °C. It is clear from Fig. 5 that irradiated CR-39 samples show no detectable fading effect with respect to the absorbed gamma dose when they were stored at room temperature within the studied fading periods indicated in the figure.

It can be said that this property of CR-39 foil (no gamma loss when stored at room temperature for about 8 days) reflects its great stability; CR-39 foil is therefore highly recommended for use as a gamma-ray dosimeter. Experimental results of V_{BD}/V_{BO} measurements as a function of fading time were also determined at 135 °C. Fig. 6a and b show the fading behaviour of gamma-irradiated CR-39 samples studied

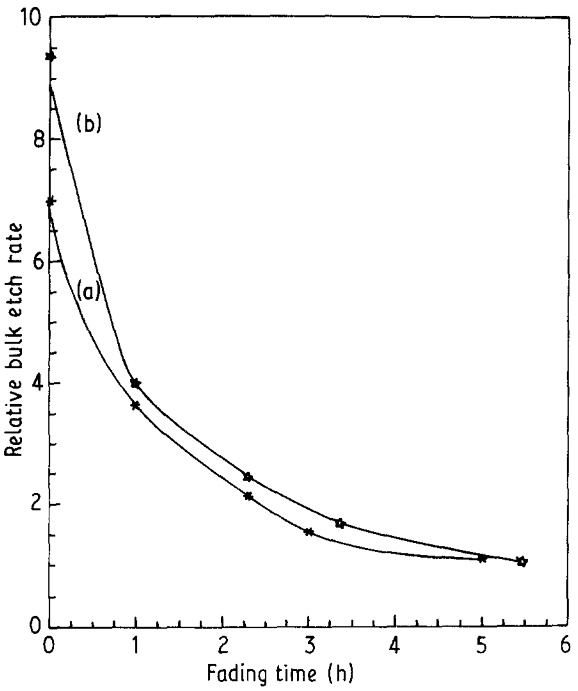

Figure 6 Effect of fading time at 135 °C on the relative bulk etch rate V_{BD}/V_{B0} measurements of CR-39 detectors, etched in 6.25 N NaOH; samples preirradiated at (a) 9 and (b) 12 Mrad.

at 135 °C for various times; the V_{BD}/V_{B0} ratio is plotted versus time for samples preirradiated to (a) 9.0 and (b) 12 Mrad. Samples were etched in 6.25 N NaOH at 70 °C. From Fig. 6, it can be noticed that at a storage temperature of 135 °C, the V_{BD}/V_{B0} value is time dependent; it decreases with increasing time and after about 5 h the effect of gamma radiation on the plastic material was almost lost.

Finally, it may be concluded that the dose-effect linearity (see Figs 1–3) and the fading data given in Fig. 5 indicate the feasibility of using the CR-39 plastic material as gamma ray dosimeters under optimum etching conditions of 6.25 N NaOH solution at 70 °C where plastics showed a linear response over a wide range of gamma dose (3–20 Mrad).

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