MC simulation of a PGNAA system for on-line cement analysis

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Abstract A prompt gamma neutron activation analysis system with a ^{252}Cf neutron source for on-line cement analysis has been simulated with the MCNP code. The results indicate that the optimum arrangement is a Bi shield of 20-mm thickness, a polyethylene moderator of 50-mm thickness, a source-to-sample distance of 70 mm, and cement samples of 1200 mm×600 mm×170 mm. To absorb thermal neutrons and suppress low-energy γ-rays, the optimum-sized sheets are 150 mm×7 mm Cd, and 150 mm×15 mm Pb.

Key words Monte Carlo simulation, Prompt γ neutron activation analysis, Moderator material, Neutron flux

1 Introduction

A prompt gamma neutron activation analysis (PGNAA) system, a non-destructive multi-elemental analysis method of high sensitive, is widely used for industrial applications[1–3]. Many elements can be determined by thermal neutron capture and neutron inelastic scattering[1], but it takes time, and costs a lot, to design an experimental installation according to neutron source specifications. PGNAA simulation can be a good solution in this regard. For example, the PGNAA instrument in analyzing cement raw materials was optimized by the MCNP code[1], and the results were compared with those obtained using 2.8 and 14 MeV neutrons from a ^{252}Cf source[2]. The lime/silica ratio in concrete could be measured[3]. Zhang *et al*[4] designed an on-line neutron activation analysis system using the MC. Yang *et al*[5] simulated key parameters of an on-line PGNAA system.

In this paper, the MCNP code is used to optimize PGNAA parameters in cement analysis by simulating the neutron transport process in the source chamber and moderators, with the neutrons in average energy of 2.3 MeV from a ^{252}Cf neutron source. The optimum type and thickness of the source shield, and the moderator material, and thicknesses of the cement sample and the Cd and Pb sheets are found. The content of Ca, Si, Fe, and Al are evaluated in sample.

2 The simulation model

The cement sample was of 2.25-g·cm^{-3} density, and the contents were: 33.88% SiO_2, 2.42% Al_2O_3, 1.77% Fe_2O_3, 0.23% Fe_3O_4, 9.4% CaO, 45.11% $CaCO_3$, 0.62% MgO, 0.45% SO_3, 0.34% K_2O, 0.12% Na_2O, and 5.66% H_2O.

Fig.1 shows the simulation model in outer dimension of 1200 mm×1200 mm×900 mm, with a source chamber of 600 mm×600 mm×300 mm, and a sample chamber of 600 mm×1200 mm×300 mm (the cement are on a product conveyer). The ^{252}Cf neutron source was enclosed by a spherical lead cavity of 10-mm radius to absorb γ-rays emitted by the source. Thickness of the moderator material around the sphere and the sample thickness were variable. The NaI(Tl) detectors were covered by Cd to adsorb thermal

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neutrons, and Pb to adsorb γ-rays below 1.5 MeV. ^{252}Cf decays in spontaneous fission with a half-life of 2.64 years, emitting neutrons in average energy of 2.3 MeV at rate of $2.3\times10^{12}\ s^{-1}\cdot g^{-1}$. This can be simulated by the MCNP code using Maxwell fissile spectrum model[6],

$$f(E) = CE^{1/2}\, e^{-E/\alpha} \quad (1)$$

where, E is the neutron energy, α=1.42 MeV corresponds to a nuclear temperature at the neutron emission, and C is a coefficient.

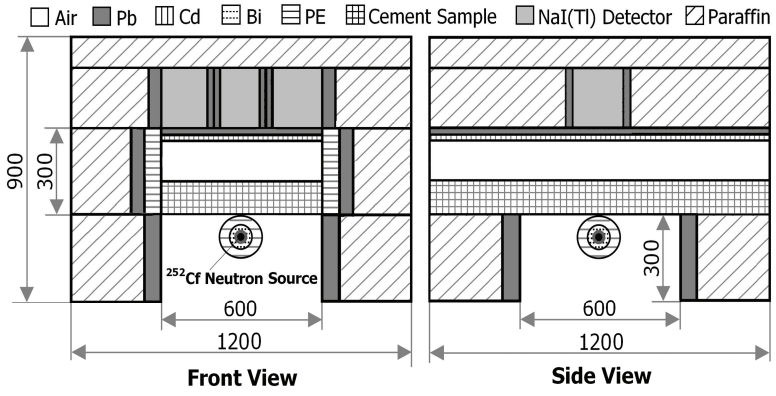

Fig.1 A schematic of the simulated setup.

3 Results and discussion

The simulation requirements included the assumption of a Maxwell fissile spectrum, a cross-section database of ENDF/B-VI Rel.1, Intel(R) Core(TM) 2CPU T7200 @2.00 GHz computer, the scored F2 tally, relative counting rates (one neutron count per unit area) for thermal neutrons (E < 0.5 eV)[7], epithermal neutrons (0.5 eV–1 keV), and fast neutrons (E >1 keV). A total number of 2×10^7 was used for a better simulation statistics. The computer time was about 2 h for each data point, at the counting errors of less than 1%.

3.1 Type and thickness of source shield materials

The γ-rays of certain energy and intensity are emitted by the ^{252}Cf spontaneous fission. To reduce the γ-ray influence on PGNAA analysis, Pb, Bi, W and Cu, as the neutron-source shield materials, were simulated, and their shielding effects are shown in Fig.2.

In Fig.2(a), the four materials behave similarly in reducing the neutron flux, which decreases rapidly below 10-mm shielding material thickness, where it begin to decrease gradually.

From Fig.2(b), W and Cu are not good because of the high γ-ray fluxes, especially with thinner than 30 mm of W or 40 mm of Cu. For Pb and Bi, the γ-ray fluxes are small, and they decrease all the way with increasing thickness.

Comparing the Pb and Bi curves, at 20–30 mm thickness, the γ-ray relative flux decrease of Bi is larger than that of Pb. Therefore, the Bi of 20-mm thickness is the best shielding material.

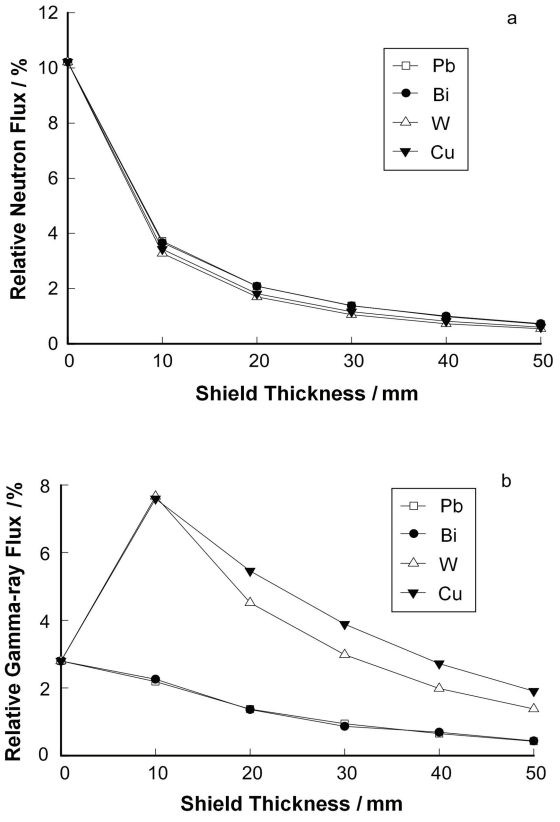

Fig. 2 Simulated neutron (a) and γ-ray (b) fluxes as function of thickness of different shield materials.

3.2 Type and thickness of the moderator materials

To ensure accurate PGNAA online analysis, the thermal neutron flux should be maximized, and the fast neutron flux minimized. Therefore, it is crucial to slow down the fast neutrons into thermal neutrons by a moderator material. Its performance is evaluated by the moderation ability and ratio, and thermal factor.

The moderation ability is defined as $\xi\Sigma_s$, where $\xi = \ln(E_0/E) = 1+[(A-1)^2/2A]\ln[(A-1)/(A+1)]$ is the energy curtailment, i.e. how much an average neutron loses its energy in each collision with the moderator atoms; A is mass number of the moderator material; and Σ_s is cross-section of macroscopic scattering. The greater the Σ_s is, the greater the probability of neutrons scattered by the moderator material. Thus, the $\xi\Sigma_s$ reflects the material's ability to moderate neutrons.

The moderation ratio is defined as $R_m=\xi\Sigma_s / \Sigma_a$ [5,8], where Σ_a is macroscopic absorption cross-section of the moderation material.

The thermal factor is TF (cm^2)$=Y_f/Y_{th}$, where Y_f is fast neutron yield(n·s^{-1}), and Y_{th} is peak thermal flux (n·cm^{-2}·s^{-1})[5,8], and the smaller the TF value, the greater moderator efficiency of the material.

The neutrons can be absorbed by the moderator, too. So, a good moderator material should have large moderation ability and ratio, but small thermal factor. Parameters of light water, heavy water, paraffin and polyethylene (PE) as moderator materials are listed in Table 1[4,5,7–10].

Table 1 Characteristic parameter values of several moderator.

Materials	Density / g·cm^{-3}	ξ	$\xi\Sigma_s$ / cm^{-1}	R_m
Light water, H_2O	1	0.92	1.35	71
Heavy water, D_2O	1.1	0.51	0.176	5.67
Paraffin, $C_{30}H_{62}$	0.9	0.94	1.705	64
Polyethylene, C_2H_4	0.96	0.91	1.8	64

With 20-mm thick Bi as source shield material, simulations were carried out on the four moderators in Table 1. The thermal and fast neutron fluxes are related with the moderator thickness. For the light water, paraffin and PE of thinner than 50 mm (Fig.3a), the thermal neutron fluxes increase with the thickness, whereas for heavy water of < 120-mm thickness, the thermal neutron flux is lower than the other materials. From Fig.3(b), the fast neutron fluxes decrease with increasing moderator thicknesses, with heavy water being the most capable of reducing a commensurate relative flux. In comparison, the thermal neutron flux by paraffin or PE changed linearly, reaching a maximum at 40–60 mm thickness, and 50-mm thick PE is the best of all moderators in this study.

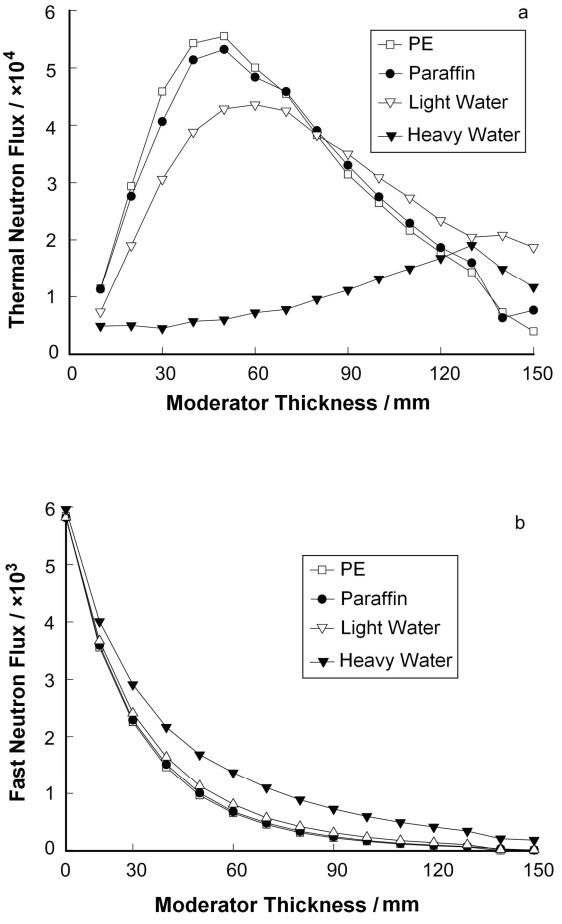

Fig.3 Thermal (a) and fast (b) neutron fluxes as a function of thickness of different moderator materials.

3.3 The source-to-sample distance

Optimizing the source-to-sample distance was realized by simulating the maximum thermal, epithermal and fast neutron fluxes, using cement samples and Bi shielding of 20 mm and PE moderator of 50 mm. Fig.4 shows that the neutron fluxes decreases slightly with increasing distance from neutron source to sample, hence the best distance of 70-mm.

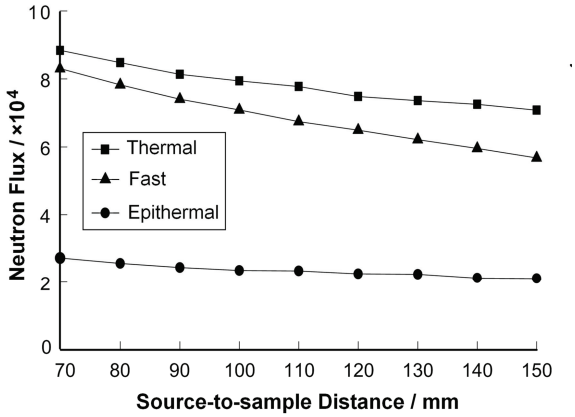

Fig.4 Flux of thermal, epithermal and fast neutrons, as a function of the source-to-sample distance.

3.4 The sample thickness

Qualitative elemental analysis of a sample can be done by detecting prompt characteristic γ-rays from the nuclides activated by thermal neutrons. The γ-ray yields were simulated with Ca, Si, Fe, and Al (the target elements of cement), through reactions (the γ-ray energies are given in MeV in the parentheses) of ^{40}Ca(n,γ) (1.942), ^{40}Ca(n,γ) (6.420), ^{28}Si(n,γ) (3.539), ^{28}Si(n,γ) (4.934), ^{55}Fe(n,γ) (7.631), and ^{27}Al(n,γ)(7.724), with abundance of the nuclides being 100%, 49%, 100%, 93%, 100%, and 96%, respectively.

The shielding and moderator were the same as in Section 3.3. In Fig.5, the characteristic γ-ray yields from Ca, Si, Fe, and Al targets of different thicknesses have been normalized to the neutron number of simulation. For Ca, the maximum yields of the 1.942 and 6.420 MeV γ-rays are at 170 and 150 mm (Fig.5a), respectively. The 3.539 and 4.934 MeV γ-ray yields of Si maximize at 80 and 110-mm (Fig.5b), respectively. The 7.631–7.646 MeV γ-ray yields of Fe in the 160–190 mm region form a plateau over other data points (Fig.5c). And the 7.724 MeV γ-ray yield of Al peaks at 190 mm (Fig.5d).

On the other hand, cement samples in thickness of 100–180 mm are practically used for elemental analysis to attain various γ-rays fluxes. In this study, we used 170 mm as the optimum sample thickness.

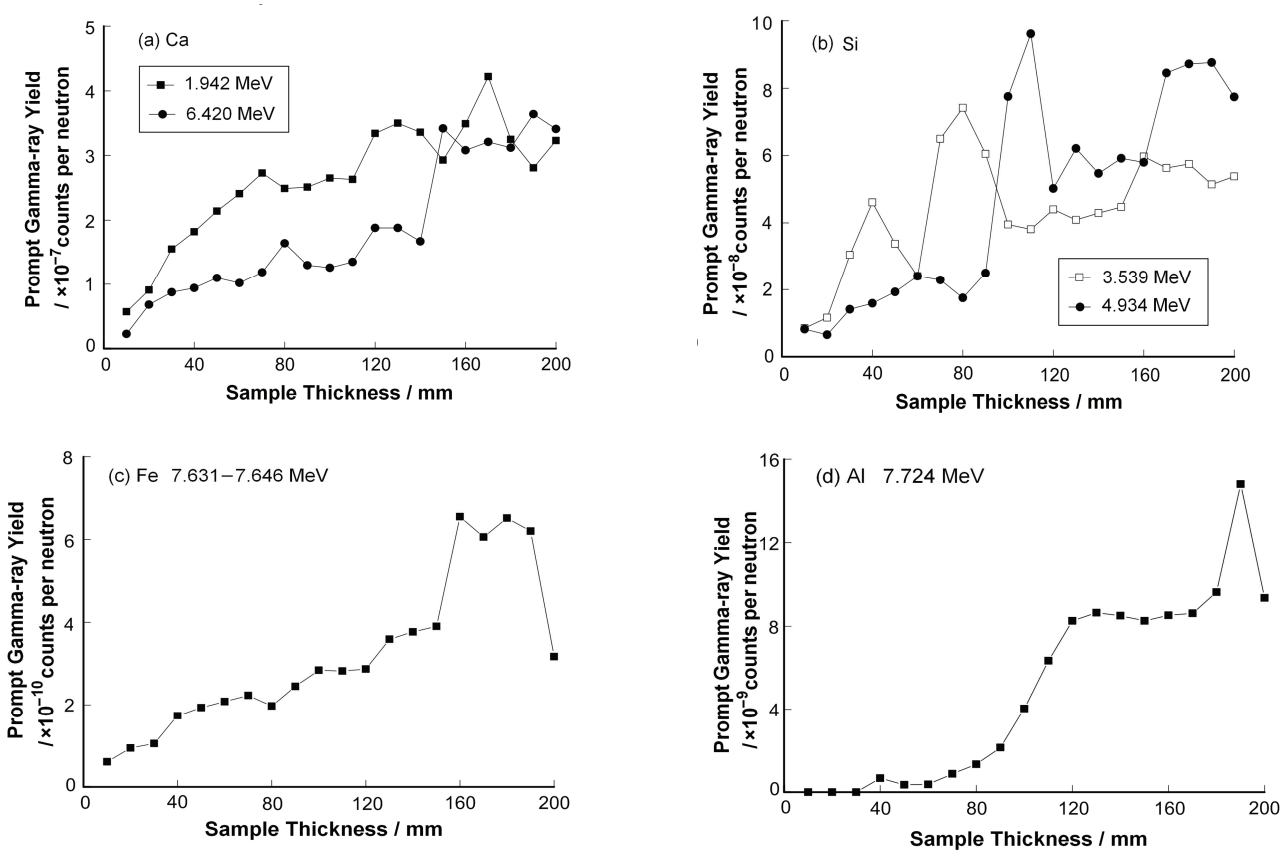

Fig.5 Normalized yields of prompt γ-rays calculated with elemental targets of different thicknesses.

3.5 Thickness of the Cd and Pb sheets

Thermal neutrons can be produced inside the sample by side reactions of (n,n), (n,n′) and (n,2n) during the (n,γ) reactions, and a Cd sheet is placed before the NaI(Tl) detectors to keep them free from the thermal neutrons. On the other hand, as energies of the γ-rays to be detected are over 1.5 MeV, a Pb sheet should be used to stop γ-rays below 1.5 MeV. The Cd and Pd thickness were simulated with a cement sample of 170 mm, a Bi shield of 20 mm, and a PE moderator of 50 mm. In Fig.6(a), the fluxes of thermal neutrons (Φ_1) and γ-rays (Φ_2, < 1.5 MeV; Φ_3, ≥ 1.5 MeV) increase with Cd thickness. The Cd sheet can stop ~80% of the thermal neutrons, 20–40% of the low energy γ-rays, and 5–20% of the high energy γ-rays. As shown in Fig. 6(b), at 7 mm of the Cd thickness, $\Delta\psi=\Phi_1-\Phi_2$ is at the minimum, and $\Delta\theta=\Phi_2-\Phi_3$ is at the maximum. This is appropriate for an optimum design.

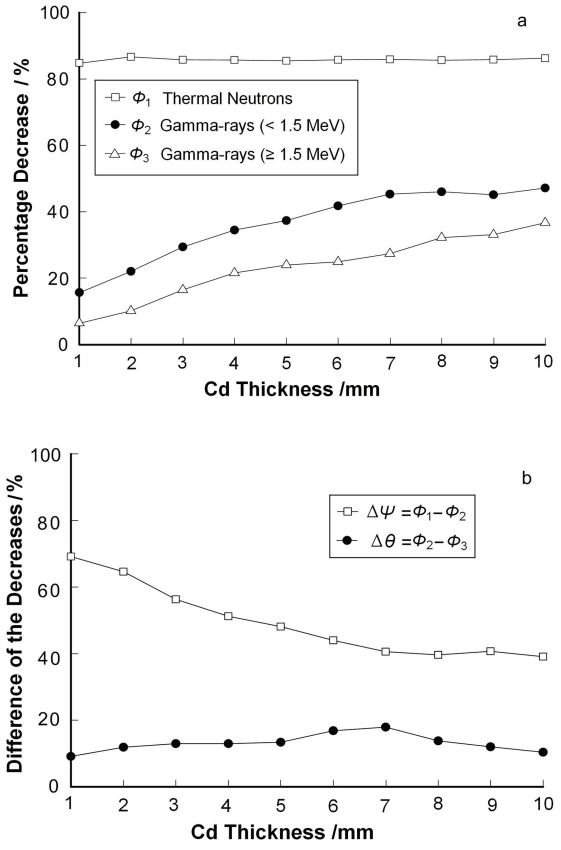

Fig.6 Percentage decrease of the thermal neutron and γ-ray flux and difference of the decreases *vs* thickness of Cd sheet.

Fig.7 shows effect of the Pb sheet on γ-ray flux. The low energy γ-rays (Φ_2) are suppressed to 10–20% and the high energy γ-rays (Φ_3) to 10–30%. The Φ_2 remains low flat from 6 to 15 mm, and the Φ_3 has a slight fluctuation. Both become higher when the Pb sheet is thicker than 15 mm, which are suitable to maximize the high energy γ-ray yield.

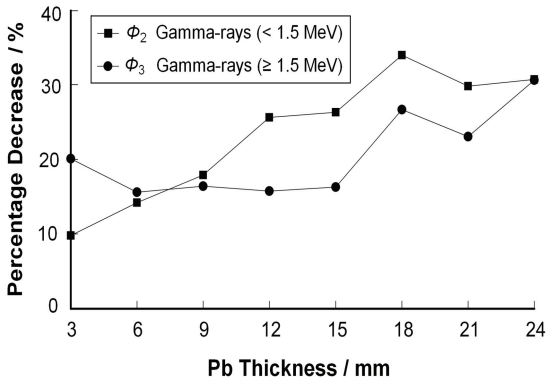

Fig.7 Gamma-ray yield decrease *vs* thickness of Pb sheet.

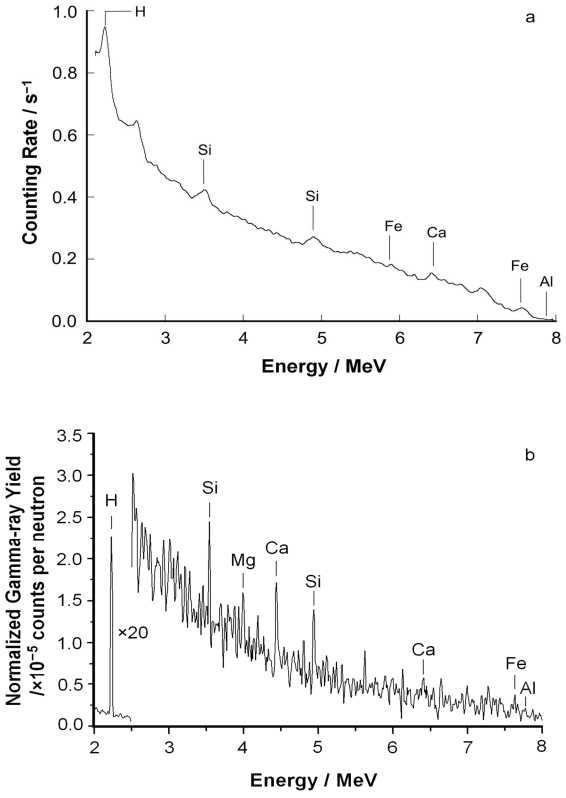

Fig.8 The measured (a) and simulated (b) spectra of prompt γ-rays from a cement sample.

4 The experimental set-up

An experimental set-up, with a 3"×3" NaI(Tl) detector, a multi-channel analyzer and software, was established based on the MC simulation results. The prompt γ-ray spectrum measured with a cement sample (Fig.8a) is in agreement with that of the MC simulation in

Fig.8(b). The characteristic γ-ray peaks are: H 2.23 MeV; Si, 3.54 and 4.93 MeV; Ca, 4.42 and 6.42 MeV; Fe, 5.92 and 7.63 MeV; and Al, 7.72 MeV. The Ca, Fe and Si content are obtained.

5 Conclusions

PGNAA on-line analysis of cement samples were simulated with the MCNP code. The optimal type and thickness of source shield and moderator materials, the sample size, and the Cd and Pb thickness, were verified with an experiment set-up using a point source and single detector. The performance should be improved by using a distributed neutron source, a larger detector of 6"×6", and Bi or Pb-Bi alloy to reduce γ-ray background.

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