# ALL-OPTICAL HARD X-RAY SOURCES AND THEIR APPLICATION TO NUCLEAR ENGINEERING

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#### Abstract

K-edge densitometry using monochromatic hard X-ray beams, which is one of the effective techniques to measure the concentration of nuclear materials in reprocessing solutions, is investigated in this study. The inverse Compton scattering between an infrared (IR) laser beam of 800 nm and high-energy electron bunch of 74 MeV makes it possible to deliver tunable monochromatic X-rays near the Kabsorption edges of nuclear materials of 115-129 keV. The only way to realize a compact densitometry system is to use a laser wakefield accelerator (LWFA) instead of a radiofrequency linac. The preliminary experiment and simulation results on the electron injection suggest that a stable LWFA, stabilizing the requirements can be realized using electron transport optics. The result of a 1-D particle-incell (PIC) simulation suggests that the characteristic density scale length of the density down ramp is about twice the plasma wavelength, and the density ratio between both sides of the density ramp is about 4.

## **INTRODUCTION**

From the viewpoints of ensuring accountability and maintaining safeguards, the concentration of nuclear materials and minor actinides (MAs) has to be measured precisely and accurately during the reprocessing of spent nuclear fuels. Hybrid K-edge/X-ray fluorescence densitometry (HKED) has been applied to measure the concentration of nuclear materials such as uranium (U) and plutonium (Pu). HKED achieves a precision and accuracy of 0.2% for U and 0.75% for Pu when the concentration of the analyte ranges from 50 to 150 g/L in a measurement time of 1000 s [1]. However, it is difficult to measure the concentration of minor elements in mixed solutions because HKED does not have a wide dynamic range [2]. Since chemical separation processes are not used in the advanced fuel cycle for Pu and MAs, a new measuring technique is required for evaluating the concentration of U, Pu, and MAs in mixed solutions. Moreover, the measurement time of the next-generation densitometer must be about 1/10th of the present one for dealing with a large amount of the spent fuels.

Intense monochromatic hard X-rays can be applied to measure concentrations. Monochromatic K-edge densitometry (MKED) using tunable monochromatic hard X-rays makes it possible to determine the concentration of all elements in a solution by measuring the transmission rate of X-rays at both sides of the K-edge of the target element in the solution. Recently, the development and application of tunable monochromatic hard X-ray sources based on the inverse Compton scattering process have been intensively researched. Superior characteristics of the inverse Compton X-ray sources, such as monochromaticity, tunability, polarization, directionality, and a short pulse, are suitable for K-edge densitometry. Because K-edge energies of nuclear materials and MAs are in the range of 115 - 129 keV, the electron energy required for producing hard X-rays tuned to K-edge energies, produced by the inverse Compton process using an infrared (wavelength: 800 nm) laser pulse, must be higher than 72 MeV.

Radio-frequency linacs are widely used to accelerate electrons to around 100 MeV; however, they are too large to be used in a reprocessing plant. Alternatively, laser wake-field accelerators (LWFA) can be used to realize a tabletop Compton hard X-ray source, i.e., an all-optical Compton hard X-ray source. This is possible because a wakefield, which has a high acceleration gradient of 1 GeV/cm, is generated by an intense laser pulse.

# ALL-OPTICAL MONOCHROMATIC K-EDGE DENSITOMETRY

## Parameters of X-rays, electron beams and LWFA

The concentration of an element in a given sample is evaluated by a simple formula:  $\rho = \ln (A_1/A_2) / (\mu_2 - \mu_1) / L$ , where A and  $\mu$  are the X-ray transmission rate and mass attenuation rate, respectively; subscripts 1 and 2 indicate the monochromatic X-ray energies of the upper and lower sides of the K-edge, respectively; and L is the thickness of the sample.

All-optical MKED for use in the advanced fuel cycle must measure element concentration with the accuracy of 0.2% for Pu within the measurement time of 100 s. Parameters of X-Rys, electron beams, and LWFAs are summarized in Table 1. In order to achieve an accuracy of 0.2% for Pu, the energy fluctuation of the X-ray photon must be smaller than 0.1% (0.1 keV); this is because the mass attenuation rate is a steep function of the X-ray photon energy. Since the K-edge energies of elements differ by 3 keV, the energy width of the monochromatic X-ray must be smaller

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	X-ray	Electron Bunch	Self- Trapping LWFA
Energy	115 – 129	$\geq 4 \; \mathrm{MeV}$	pprox 100  MeV
Range	keV		1mm long
Energy	< 0.2%	pprox 0.1%	10 - 20 %
Fluctuation	(0.23keV)	(0.1keV)	(*1)
Energy	< 3  keV	< 1.5  keV	5 - 20 %
Width	( $\approx 2.3\%$ )	$(\approx 1.2\%)$	(*1)
Number of Particles /shot (100 Hz)	> 10 <sup>5</sup> photons /3 keV/shot	$\approx 100 \text{ pC/shot} \\ (6 \times 10^8 \\ \text{electrons})$	$\approx 10 \text{ pC}$

Table 1: Required and Present Parameters.

\*1. These can be relaxed by using proper electron transfer optics.

than 3 keV ( $\leq 3\%$ ).

The number of X-ray photons is estimated to be  $10^5$  from the HKED data of the reference [2]. Since the number of Xray photons at the K-absorption edge after passing through a 2-cm thick sample containing 100g/L U was about  $10^8$ photons/keV/1000s, the number of incident photons was estimated to be approximately  $3 \times 10^8$  photons/keV/1000s. The minimum number of photons required for measuring the concentration in 100 s is  $10^9$  photons/3 keV. Then, the number of photons emitted from the inverse Compton Xray source in one shot is estimated to be  $10^5$  photons/3 keV/shot, where the repetition rate is assumed to be 100 Hz. The electronic charge required to produce  $10^5$  photons by inverse Compton scattering at the IR laser intensity of  $10^{17}$ W/cm<sup>2</sup> is approximately 100 pC.

The right most column of Table 1 lists typical parameter values of an LWFA operated in the self-trapping mode of initial electrons. Although the energy fluctuation, energy width, and electronic charge of the self-trapping LWFA are far from the required values, these can be improved to satisfy the requirements by adopting the forced injection of electrons into the LWFA and suitable electron optics to focus electrons at the collision point.

# Rough Design of All-Optical MKED System for Measuring Concentration of Nuclear Materials

The all-optical MKED system consists of a terawatt (TW) pulse laser, a focusing mirror, a small hort plasma channel, an electron lens, an X-ray collimator, a sample, and an X-ray detector, as shown in Fig. 1. The X-ray energy is determined by Eq.1, where  $\hbar\omega_L$ ,  $\gamma$ ,  $\phi$ , and  $\theta$  are the photon energy of the laser, electron energy normalized by the electron rest mass, crossing angle (collision angle), and scattering angle from the beam axes of the electrons,



Figure 1: Schematic drawing of all-optical MKED system.

Table 2: X-ray photon energies and crossing angles required to measure concentration of elements.

Elements	K-Absorption Edge Energy (keV)	X-ray Energy (keV)	Crossing Angle (°)
		114.07	41.6
U	115.60		
N	110 (7	117.14	37.4
Np	118.67	120.20	22.0
D11	121 70	120.29	32.8
I u	121.79	123.39	27.1
Am	124.98	120107	
		126.61	20.0
Cm	128.24		
		129.87	8.2

Electron energy = 74 MeV, Laser wavelength = 800 nm.

respectively.

$$E_X = 2\hbar\omega_L\gamma^2 \left(1 + \sqrt{\gamma^2 - 1}/\gamma\cos\varphi\right) / \left(1 + \gamma^2\theta^2\right)$$
(1)

The empirical scaling law of the self-trapping LWFA shows that the laser power and the plasma density required to accelerate electrons up to about 100 MeV are about 12 TW and  $\leq 1.5 \times 10^{19} {\rm cm}^{-3}$ , respectively [3]. The number of accelerated electrons obtained by extrapolating the experimental data on the laser power dependence of the electronic charge is  $10^9$  [4]. The focal length of an off-axis parabolic mirror is chosen in the range of 150 to 250 mm for focusing the laser pulse to an intensity of  $10^{19} {\rm W/cm^2}$ .

The optimum length of the plasma channel is chosen to be equal to the dephasing length of electron acceleration, which is approximately 1 mm for the plasma density of  $1.5 \times 10^{19} {\rm cm}^{-3}$ . Since the typical divergence angle of the

electron beam from the LWFA is 17 mrad (FWHM), an electron lens must be used to focus electrons at the collision point to increase the electron density. The tolerable focusing angle of the electron beam, which affects on the broadening of the X-ray spectrum, is estimated by differentiating Eq. 1. The focusing angle of the electron beam should be less than 70 mrad ( $4^\circ$ ) to avoid the extra broadening of the X-ray spectrum. Therefore, the magnification ratio of the electron focusing lens of about 1:1 is suitable for the all-optical MKED. The use of an electron focusing lens made of a quadrupole magnet, which has an ultra-high field gradient of 300 T/m, makes it possible to construct a compact electron transport system [5].

X-ray photon energies and crossing angles required to produce X-rays for the measurement of the concentration of nuclear materials and MAs are listed in Table 2. The angle interval of each collision laser beam of around 5° restricts the F-number of the focusing optics. The focusing optics with the minimum F-number of F/12 is sufficient for producing a long Rayleigh range of 140  $\mu$ m at the collision point.

A collimator made of a 3- to 5-mm-thick tantalum or lead sheet is used to select an appropriate energy region of the X-ray beam with the energy width of less than 3 keV, which is placed at the distance of 10 cm from the collision point. The diameter of the collimator hole is less than 0.1 mm.

The coincidence detection of X-rays triggered by the collision laser pulse is effective for reducing the noise level in nuclear fuel reprocessing plants.

## Improving LWFA

In order to improve the energy stability, energy width, and electronic charge of the output of the LWFA, injection of initial electrons, amplitude of the wakefield, and acceleration length must be well controlled. The establishment of the injection technique of initial electrons is essential for developing a practical LWFA. Two injection techniques, optical injection [6] and wavebreaking at a density ramp of the plasma [7], have been proposed and studied. The optical injection of electrons into the wakefield has been successfully demonstrated [8]. However, it is difficult to achieve an accurate collision of two laser pulses in a plasma channel. In order to obtain an initial electron bunch by a simple process, experiments on electron injection by wavebreaking in the density down ramp, which is produced by an oblique shock wave in a supersonic gas jet, and 1-D and 2-D particle-in-cell (PIC)-simulations were carried out. The simulation results (Fig. 2) revealed the optimum conditions. The optimum density scale length and the density ratio between both sides of the density ramp are about twice of the plasma wavelength and 4, respectively. The preliminary experiment suggests an effective injection of electrons into the wakefield [9]. However, the observed electronic charge was lower than the expected one because of the machining error of the supersonic nozzle.



Figure 2: Results of 1-D PIC simulation; (a) model distribution of plasma density; (b) injected electronic charge, and (c) energy width as a function of characteristic length of density ramp.

#### **SUMMARY**

All-optical MKED is suitable for measuring the concentration of nuclear materials and MAs in nuclear fuel reprocessing plants. The required electron energy, energy fluctuation, and energy width are 74 MeV, 0.1 keV, and 1.5 keV, respectively. Although, the performance of the present LWFA in the self-trapping mode is below the required level, the preliminary experiment and simulation results suggest that the LWFA can be improved by injecting initial electrons, and using suitable electron transport optics. The simulation result suggests that the characteristic density scale length of the density down ramp is about twice the plasma wavelength, and the density ratio between both sides of the density ramp is about 4.

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