방사선방어학회지
제10권 제1호 1985

Development of Monoenergetic Photon Source in the Energe Range below 100 keV by the X-ray Fluorescence Method

Youn-Myoung Lee, Kun Jai Lee

Korea Advanced Institute of Science and Technology

Suck-Ho Hah, Sun-Tae Hwang, Kyung-Ju Lee

Korea Standards Research Institute

=**Abstract**=

The development of monoenergetic photon sources using K_α fluorescence X-ray of pure material was carried out in the energy range below 100 keV. The monoenergetic photons are very useful in the calibration of the radiation measuring instruments and can be produced as the K_α fluorescence X-ray by irradiating the bremsstrahlung to the thin pure metal foils called 'radiators'. In this experiment, several radiators such as $_{47}$Ag, $_{50}$Sn, $_{68}$Er, $_{70}$Yb, and $_{82}$Pb provide the wide monoenergetic photon energy ranging from 20 keV to 80 keV. By the spectrometry with HpGe LEPS, spectral purity factors which measure the monochrometicity for the K_α fluorescence X-ray, were determined as 0.64~0.94. Dosimetry for the purpose of the determination of the exposure rate with a 600cc thin window ionization chamber, which was calibrated by the standard free-air ionization chamber, was performed. Exposure rates ranging 8.3~232.5mR/h was obtained according to the K_α fluorescence X-ray energy for each radiator.

1. Introduction

Intense photon sources of low energy and high monochromaticity are very useful in many research areas such as X-ray fluorescence analysis, diagnostic application and calibration of detectors in the low energy range.

In the field of radiation dosimetry and measurement, the calibration of the detectors with photons of low energy is especially of great importance in connection with the sensitivity of detectors.

In order to provide a photon radiation field suitable for calibrating the instruments such as X and gamma ray detectors over the energy range from a few keV to about 10 MeV, it is required to prepare a complete set of X-ray equipment as well as a number of different radionuclide sources. The wide intensity range of sources is also necessary to provide the level of photon radiation field which can be varied from natural background levels up to very high levels, corresponding to thousands of roentgen per hour in terms of exposure rate in air. The discrete or monoenergetic energy is basically required since the sensitivity of the most radiation measuring instruments varies considerably with photon energy.

For photon energies above 300 keV, the radionuclide photon sources such as ^{137}Cs and ^{60}Co are usually used for the calibration, but below that energy, very few radionuclide sources have such a long enough half-lives, and in many cases, they emit photons of many different energies. In addition, radionuclide sources are generally rather limited in their availability not only because of their limited intensity range with small dimensions of source coming from self-absorption effect but also lack of available radionuclide sources. In particular, below 100 keV, those problems become much more severe. In the energy range below 300 keV, therefore, the demand of the photon sources has been filled unwillingly with the use of heavily filtered bremsstrahlung and characteristic X-ray photon spectrum which are generated in the X-ray tube of a X-ray generator. The filtered X-ray spectrum does not suffer from any drawback of limited intensity range as in radionuclide sources. However, in spite of the heavy filtration, there exists a considerable photon energy spread. Consequently, if we use the filtered X-ray photon source in an energy range where the sensitivity of radiation measuring instruments indicates strong variations, its reading may be misleading since we only know the effective energy of photon spectrum which was averaged for all energies of photons instead of their true energies. To overcome this problem, an accurate information for the photon energy should be provided. For this reason, it is frequently necessary to develop the photon sources which are considerably monoenergetic and have a wide range of photon energies.

In this respect, the K_α fluorescence X-rays produced by irradiating the radiators with the ordinary X-ray, so-called primary X-ray from the X-ray tube, should be worth of consideration for the energy range below 100 keV because of their high degree of monochromaticity. For past decades, many researches on the similar work about the K_α fluorescence X-ray in connection with the development of the monoenergetic photon sources have been performed[1~5].

The principle involved in this method is the conversion of the continuous X-ray distribution from the X-ray generator to a monoenergetic photon beam of K_α fluorescence X-ray through the well-known photoelectric effect in the radiator atom.

In the present work, combination of several parametric studies which may exhibit an important effect upon the variation of the K_α fluorescence X-ray, development of monoenergetic photon sources in the energy range from about 20 keV to 80 keV is presented.

2. Fundamental Theory on K_α Fluorescence X-ray

Fluorescence K_α X-rays are those emitted by the given radiator atoms as the atoms lose energy after being raised to the excited states due to the photoelectric effect of the primary X-ray. Practically, the predominant type of interactions involved with the incident primary X-ray for the radiator atom considered here are regarded due to the photoelectric effect.

Once an X-ray photon whose energy is greater than the critical K absorption edge energy is absorbed by an atom of radiator, a K-shell electron may be removed from the K shell of the atom, and then the atom remains in the excite state. Usually another electron from higher energy state of L, M,or N shells will take any vacancy of the emitted one and thus the atom will emit a photon which will be called as K fluorescence X-ray with energy characteristic of the difference in energy levels involved. Since the fluorescence X-ray has the energy corresponding to this difference, the energy of fluorescence X-ray can be considered as the monoenergetic photon. The K fluorscence X-ray comprises four principal lines; $K_{\alpha1}$, $K_{\alpha2}$, $K_{\beta1}$, and $K_{\beta2}$ fluorescence X-ray.

Obtaining the K_α X-ray, in other words, $K_{\alpha1}$ plus $K_{\alpha2}$ X-rays, with high purity for the energy distribution of photon is the subject of the present work. This K_α fluorescence X-ray has relatively high intensity than any other K fluorescence X-ray.

Primary X-ray photons emanating from the tungsten target in X-ray tube of X-ray generator are used to excite the atom of radiator. X-rays which shape a continuous energy spectrum have many advantages for the irradiation of the radiator. High flux of photons for the photoelectric effect can be obtained and since the energy of the X-ray which has the majority of photon numbers can be adjusted to match closely the K absorption edge, high spectral purity for K_α fluorescence X-ray is also attainable.

Once fluorescence X-ray is produced in the radiator atom, this X-ray should be filtered by a foil metal whose K absorption edge falls on between the energies of the K_α and K_β fluorescence X-rays in order to reduce the intensity of the K_β fluorescence X-ray. Then most of the K_β fluorescence and virtually all of the L fluorescence X-rays whose energy are located in low energy region compared to the K_α fluorescence are eliminated by this filtration.

The characteristics of the radiators selected for the present work are tabulated in Table 1. The materials should have a minimum purity of 99.9% according to the recommendation of ISO⁶⁾ for obtaining high spectral purty of K_α fluorescence X-ray. With these radiators, a wide range of photon energies can be obtained by appropriate choice of radiator materials corresponding to their K_α energies, i.e., appr-

Table 1. Characteristics of Selected Radiator Materials

Radiator Material		K-Edge Energy (keV)	Kα Energy (keV)	Kβ Energy (keV)	Purity(%)	Thickness (mg cm-2) (mm)
Silver	$_{47}$Ag	25.514	22.103	25.008	99.9+	48.37(.0046)
Tin	$_{50}$Sn	29.200	25.192	28.573	99.9	156.05(.0214)
Gadolinium	$_{64}$Gd	50.239	42.750	48.918	99.9	234.44(.0295)
Erbium	$_{68}$Er	57.486	48.801	55.930	99.9	308.54(.0337)
Ytterbium	$_{70}$Yb	61.332	52.014	59.652	99.9	302.30(.0431)
Lead	$_{82}$Pb	88.004	74.159	85.370	99.9	692.73(.0611)

oximate energy from 22 keV to 75 keV can be produced.

3. Experiments

The complete specification of photon sources such as X- and gamma radiation at the plane of interest must include not only the quantitative expression such as exposure rate but also the qualitative nature of the radiation. Such information is required to perform the calibration accurately.

To this end, two fundamental experiments for the determination of the energy and the intensity or dose rate of the K_α fluorescence X-ray are performed. To measure the spectral purity factors, the limiting aperture of the Ge detector is placed at 160cm midpoint of radiator which is located at 41cm from the focal spot of the X-ray tube. Primary X-ray beam size is determined by the limiting lead diaphragm of 2.9 cm in diameter, and similarly the fluorescence X-ray beam size is controlled with limiting lead diaphragm of 0.2 cm in diameter.

To determine the optimum voltage applied in the X-ray tube, spectral purity factors are measured at the interval of 20 KV. For the determination of the exposure rate, the front surface of the 600 cc thin window ionization chamber is placed at 50cm from the midpoint of radiator. The ionization chamber is calibrated by the standard free-air ionization chamber.

3.1 Spectrometry

The K_α fluorescence X-ray spectrum emanating from each radiator is measured with the low energy photon spectrometer (LEPS) which composed of several equipments. Schematic block diagram which illustrates the photon spectrometry system is shown in Fig. 1.

Special precautions were taken to ensure that both the Ge detector and the radiator are coaxial with the limiting lead diaphragm. This problem has been solved by aligning them with the aids of two He-Ne laser beams. Several lead diaphragms for the purpose of limiting and controlling the beam size and intensity are brought into the system.

A radiator holder to which radiator can be mounted is designed and fabricated based on the results of the theoretical calculations

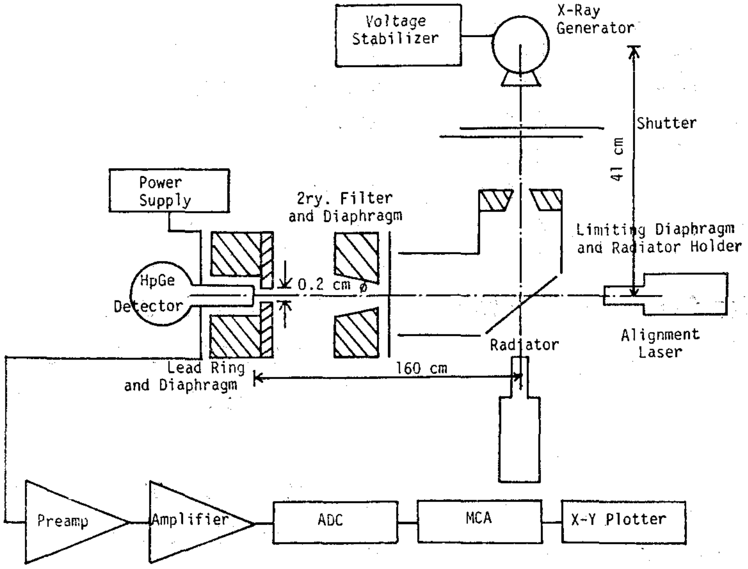

Fig. 1. Block Diagram of Spectrometry System.

with the Monte Carlo simulation[8,9] for the full system production of K_α fluorescence X-ray.

Once spectrometry siystem is prepared, the pulse height scale in multichannel analyzer (MCA) is calculated by means of standard gamma ray source to identify the various peaks in terms of spectrum.

The K_α fluorescence X-ray produced in the radiator is detected by hyperpure Ge(HpGe) detector. The spectral purity factor for K_α fluorescence X-ray is obtained by integrating the spectrum with respect to the channel number on MCA. These calibrations have been done repeatedly for several applied voltage of X-ray tube in order to determine the optimum voltage at whcih the highest spectral purity could be achieved.

Spectral purity factors for various radiators as a function of the applied voltage are represented in Figs. 2 through 7 to demonstrate that optimum applied voltage, at which maximum spectral purity can be achieved, exists. The results of photon spectrometry are listed in Table 2.

Selected spectra for various radiators are also shown in Figs. 8 and 9 which represent ${}_{68}$Er and ${}_{70}$Yb, respectively. These spectra commonly show sharp peak for a K fluorescence X-ray. Therefore, it clearly shows that, provided that the appropriate filters are used, high degree of K_α fluorescence X-ray is achievable. But spectra shown in Figs. 10 and 11 which represent ${}_{82}$Pb and ${}_{29}$Cu, respectively, show that the peak for K_β is steep persistently because of the inadequate or no filters.

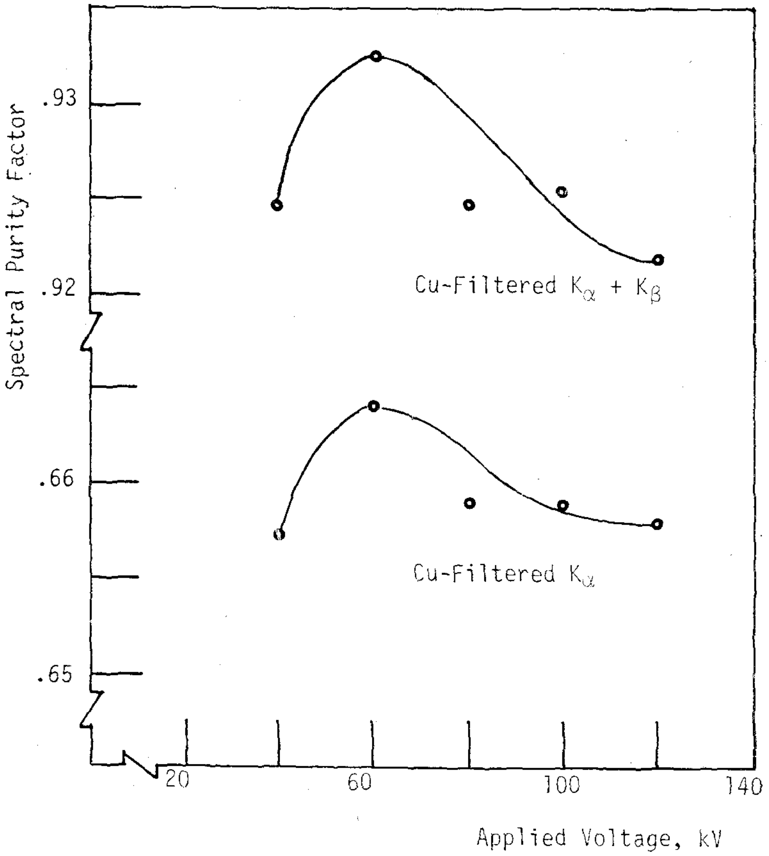

Fig. 2. Spectral Purity Factor for Radiator of Silver ($_{47}$Ag), 10mA.

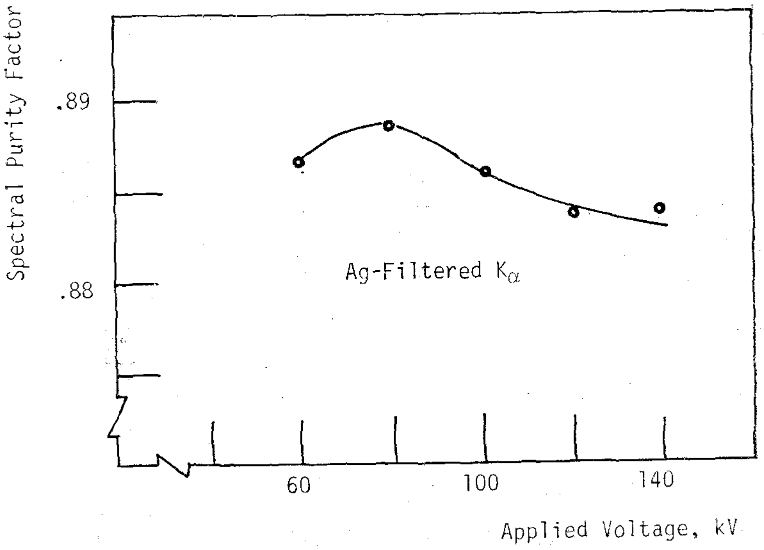

Fig. 3. Spectral Purity Factor for Radiator of Tin ($_{50}$Sn), 10mA

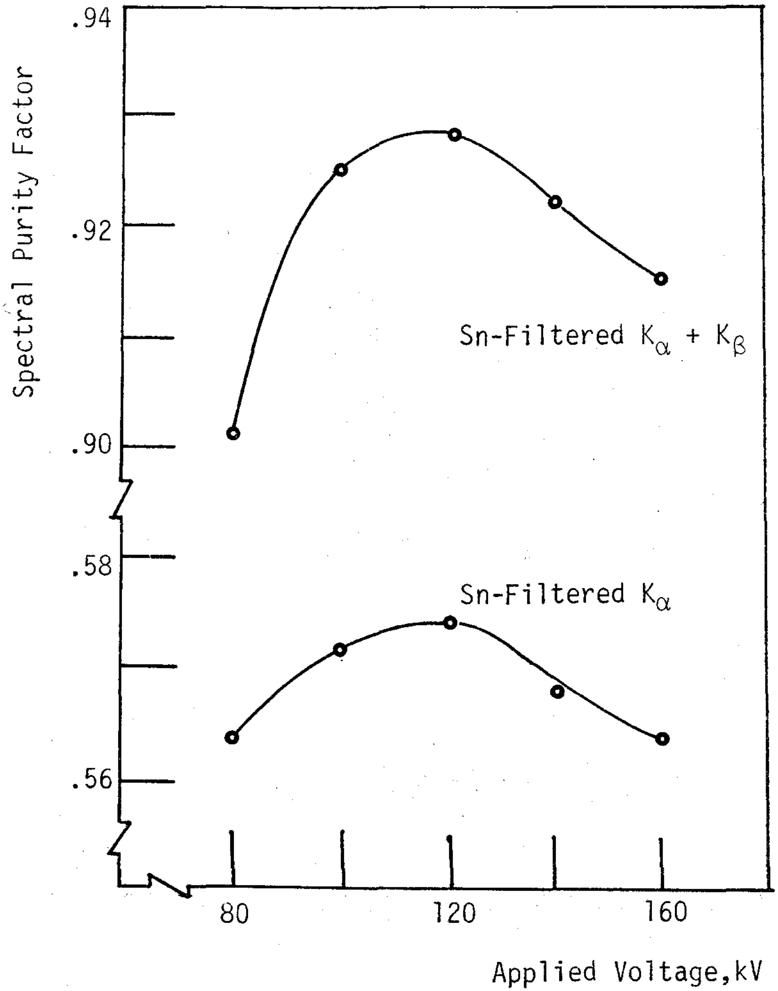

Fig. 4. Spectral Purity Factor for Radiator of Gadolinium($_{64}$Gd), 10mA.

Table 2. Results of Spectrometry at Optimum Applied Voltage for Various Radiators with Secondary Filtration

Radiator					Secondary Filter			Fluorescence X-Ray	
Material	K-Edge Energy (keV)	$K\alpha$ Energy (keV)	$K\beta$ Energy (keV)	KVP, me (KV/mA)	Meterial	K-Edge Energy (keV)	Thickness (mgcm-2) (mm)	$K\alpha$ Spec. Purity Factor	$K\alpha+K\beta$ Spec. Purity Factor
Silver $_{47}$Ag	25.514	22.103	25.008	60/10	Copper $_{29}$Cu	8.980	71.68 (.0800)	.66	.93
Tin $_{50}$Sn	29.200	25.192	28.573	80/10	Silver $_{47}$Ag	25.514	48.37 (.0046)	.89	—
Gadolinium $_{64}$Gd	50.239	42.750	48.918	120/10	Tin $_{50}$Sn	29.200	156.05 (.0214)	.57	.93
Erbium $_{68}$Er	57.486	48.801	55.930	120/10	Gadolinium $_{64}$Gd	50.239	234.44 (.0295)	.92	—
Ytterbium $_{70}$Yb	61.332	52.014	59.652	120/10	Erbium $_{68}$Er	57.486	308.54 (.0337)	.94	—
Lead $_{82}$Pb	88.004	74.159	85.370	220/10	Copper $_{29}$Cu	8.980	71.68 (.0800)	.64	.84

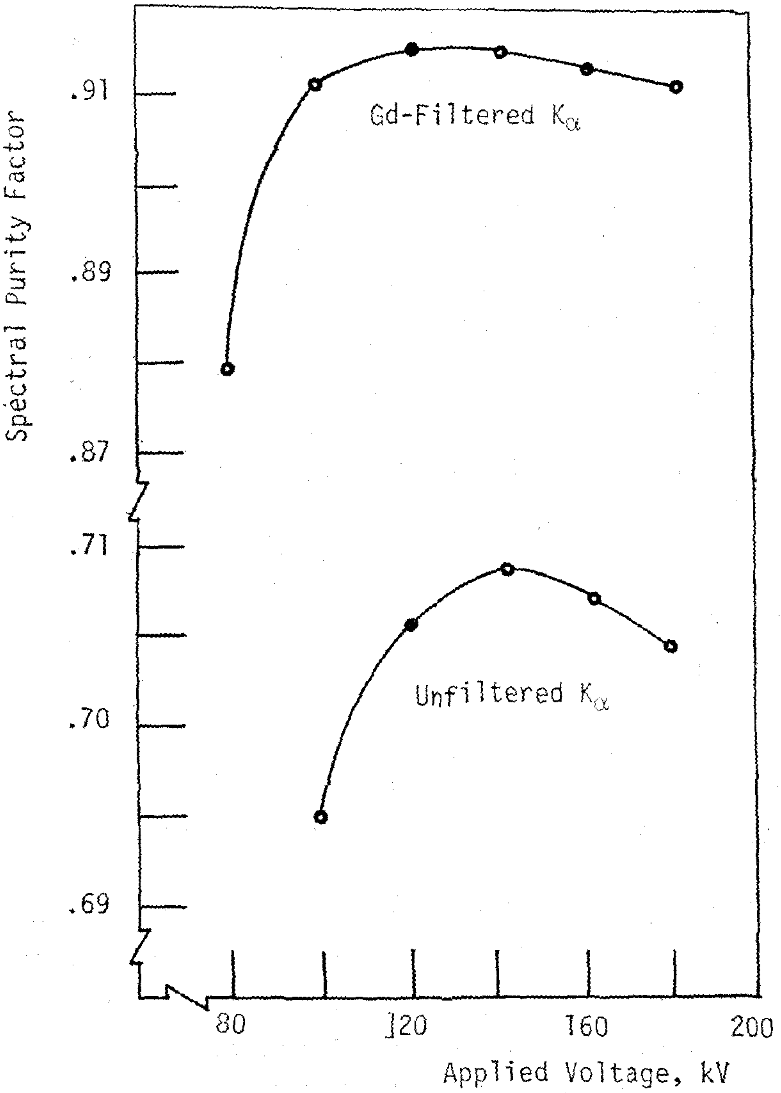

Fig. 5. Spectral Purity Factor for Radiatorof Erbium ($_{68}$Er), 10mA

3.2 Dosimetry

The fluorescence X-ray source has an elliptic form oriented 45 degree to the irradiation axis. Moreover, this source is inhomogeneous since the interactions of the flux of primary X-rays with the emitted X-ray fluorescence photons are not constant throughout the irradiated surface of the radiator. Hence, in the case of dosimetry of K_α fluorescence X-ray with the ionization chamber the error due to the inhomogeneity should be considered. Percent error resulted from considering the extended source as a point source is evaluated both on the theoretical and experimental basis. It is assumed that the 600cc ionization chamber has total surface area of 13×13cm^2 and every small area of 1×1cm^2 *in radiator*

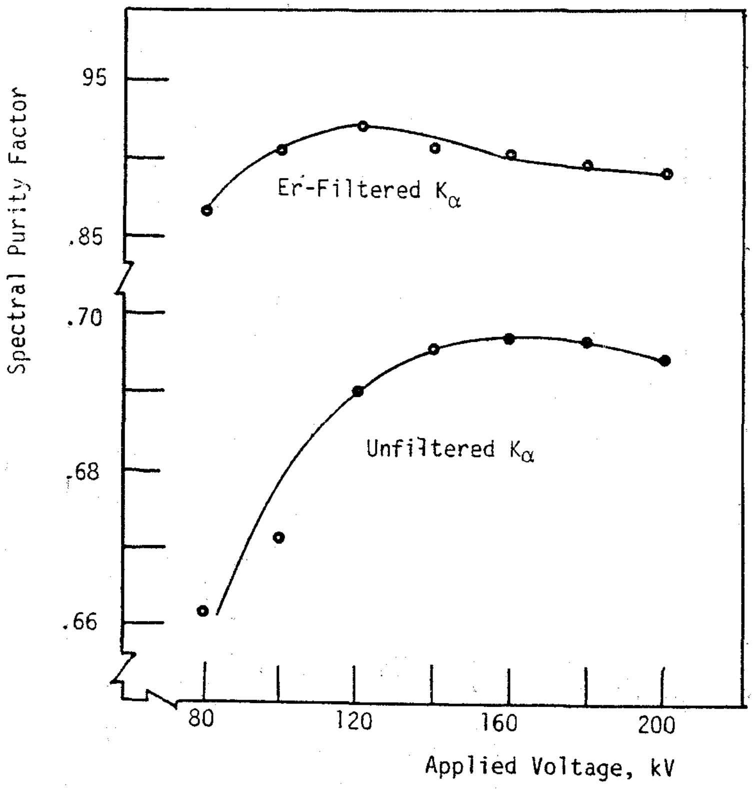

Fig. 6. Spectral Purity Factor for Radiator of Ytterbium($_{70}$Yb), 10mA

is a point source of K_α fluorescence X-ray. Analytical calculation show that homogeneous error is only 4.1 percent. Meanwhile the densitometry measurement of photographic X-ray film, which was irradiated by the K_α fluorescence X-ray, for the determination of the homogeneity of K_α fluorescence X-ray is also performed.

From these results, the 600cc thin window ionization chamber which has the window area of 100 cm² is supposed to have received full homogeneous beam of the K_α fluorescence X-ray.

Electrical charge produced in the 600cc thin window ionization chamber by the fluorescence X-ray is recorded by electrometer. With modification in terms of the temperature and pressure correction, the exposure rate expressed in miliroentgen per hour can be obtained by the following formula:

$$\dot{X}=\frac{\Delta Q}{\Delta t}\cdot S\cdot\frac{P_0}{P}\cdot\frac{T+273.15}{T_0+273.15}\cdot 3600$$

where, $\dot{X}$=exposure rate in mR/h

ΔQ=collected charge during time Δt in nC

Δt=measured time in second

P_0, T_0=standard pressure and temperature

P, T=pressure and temperature at time of measurement in mmHg and °C, respectively, and

S=2575 thin window I.C sensitivity or

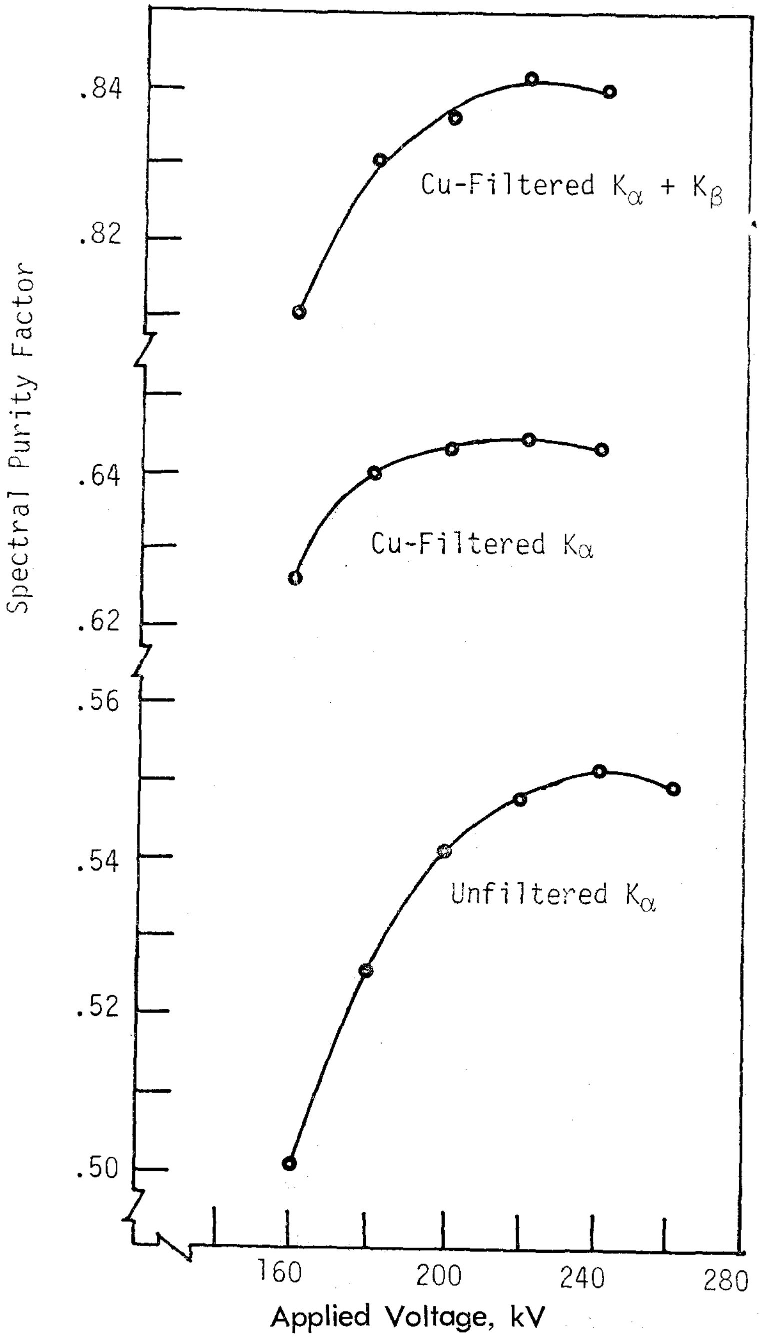

Fig. 7. Spectral Purity Factor for Radiator of Lead ($_{82}$Pb), 10mA.

correction factor for the energy measurement in mR/nC.

Sensitivity obtained by comparing the 600 cc thin window ionization chamber with FAC is a correction factor by which the electrical charge can be converted into the exposure

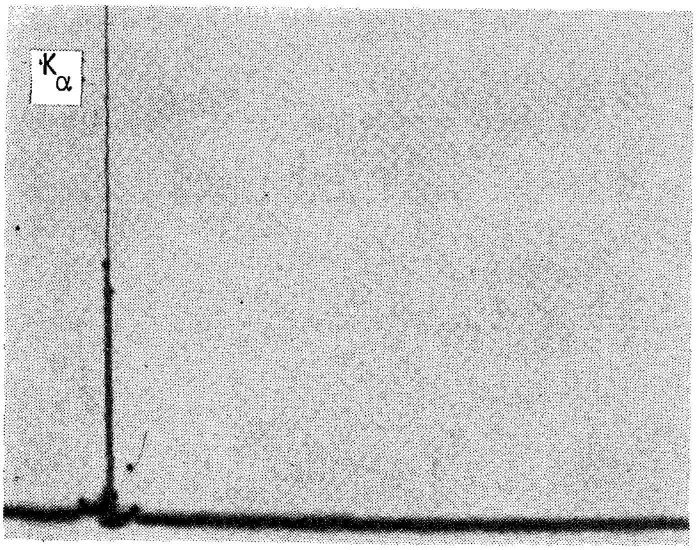

Fig. 8. Spectrum of $_{68}$Er

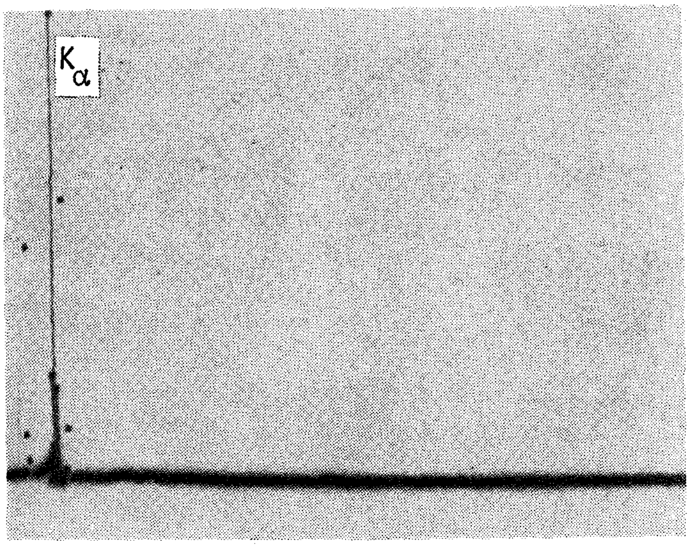

Fig. 9. Spectrum of $_{70}$Yb

rate.

4. Results and Conclusion

The spectral purity factors and the exposure rates for the K_α fluorescence X-ray of selected radiators have been measured. For some radiators such as $_{50}$Sn, $_{68}$Er, and $_{70}$Yb, relatively high degree of spectral purity factors ranging from 0.89 to 0.94 were achieved.

With selection of filter materials having adjacent atomic number, the absorption edges of the filters lie near the energy regions of K_β fluorescence X-ray of every radiator so

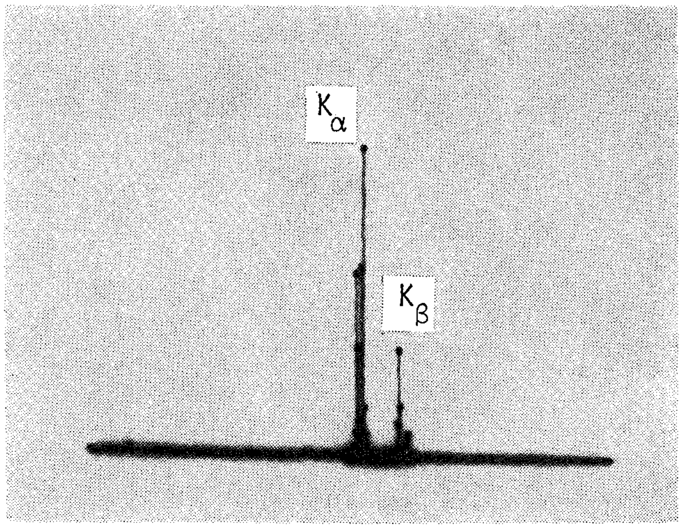

Fig. 10. Spectrum of $_{82}$Pb

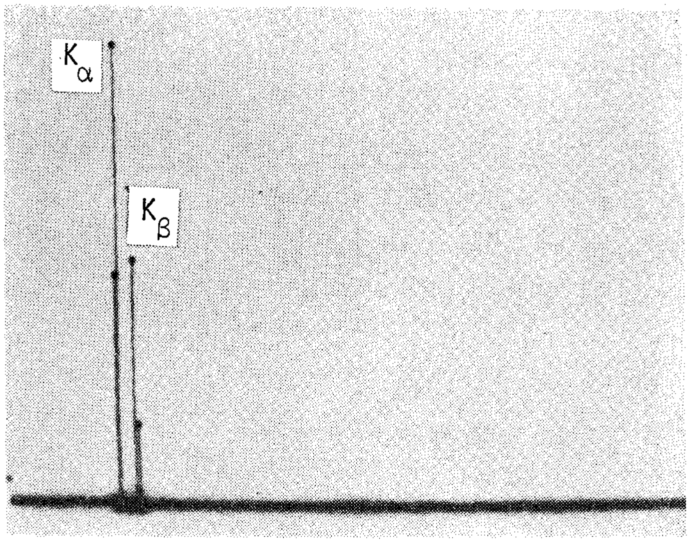

Fig. 11. Spectrum of $_{29}$Cu

that large part of K_β fluorescence X-ray from the radiator can be absorbed. For radiators such as $_{47}$Ag and $_{82}$Pb, relatively poor spectral purity factors of 0.66 and 0.64, respectively, were obtained. For these radiators, spectral purity factors for K_α plus K_β fluorescence X-ray were also obtained.

The exposure rates were determined to be 8.3~232.5 miliroentgen per hour as is shown in Table 3. For each radiator, the measurement of the charges produced in 600cc thin window ionization chamber were performed repeatedly about 15 times and the values were averaged.

Consequently, highly monoenergetic fluorescence X-ray source in the energy range from

Table 3. Results of Dosimetry for Various Radiators

Radiator (Filter)	Photon Energy (keV)	Charge (nC/30sec)	Sensitivity (RnC^{-1})	Dose Rate ($mR\cdot h^{-1}$)
Ag(Cu)	22.103	0.0135[1] (5.22×10^{-4})[2]	5.08×10^{-3}	8.3
Sn(Ag)	25.192	0.0595 (6.742×10^{-4})	5.03×10^{-3}	35.9
Gb(Sn)	42.750	0.0345 (6.742×10^{-4})	4.84×10^{-3}	19.9
Er(Gd)	48.801	0.0452 (6.742×10^{-4})	5.02×10^{-3}	27.3
Yb(Er)	52.014	0.0350 (7.303×10^{-4})	5.12×10^{-3}	21.7
Pb(Cu)	74.159	0.3420 (5.547×10^{-3})	5.65×10^{-3}	232.5

[1] Mean, $\bar{X}=\frac{\sum_i X_i}{N}$

[2] S.D., $s=\sqrt{\frac{\sum_i (X_i-\bar{X})^2}{N-1}}$

20 to 80 keV and with exposure rates up to 232.5 mR/h have been developed. These X-rays are extremely useful in the calibration of detectors as a function of energy sensitivity and for any system whose energy dependence in this energy range must be confirmed. In addition, it is expected that the fluorescence X-ray obtained in the present work should be of a great value to the establishing of radiation standards in the energy range below 100 keV.

Preparation of various different kind of radiators and adequate filters may guarantee the further useful feature of the monoenergetic photon sources.

REFERENCES

1) H.V. Larson, I.T. Myers and W.C. Roesch, "Wide-Beam Fluorescence X-ray Source", Nucleonics, Vol. **13**(11), (1955), pp. 100-102.
2) R.L. Kathren, F.L. Rising, and H.V. Larson, "K-Fluorescence X-rays: A Multi-Use Tool for Health Physics", Health Physics, Vol. **21**, (1971), pp. 285-293.
3) P.J. Ebert, J.L. Gaines, and G.R, Leipelt, "Production of Monoenergetic X-ray Sources of Known Absolute Intensity", Nucl. Inst. Meth., Vol. **99**, (1972), pp. 29-34.
4) J.L. Chartier, G. Portal, D. Roman et D. Duguay, "Production de Rayonnements Monochromatique Intenses", Nucl. Inst. Meth., **100**, (1972), 107-119.
5) E.J. Hoffman and M.E. Phelps, "Production of Monoenergetic X-rays from 8 to 87 KeV", Phys. Med. Biol., Vol. **19**(1), (1974), pp. 19-35.
6) "X and γ Reference Radiation for Calibrating Dosemetes and Dose Ratemeters and for Determining Their Response as a Function of Photon Energy", Ref. No. **ISO** 4037-1979(E), (1979).
7) N.A. Dyson, "Characteristic X-rays-A Still Developing Subject", Phys. Med. Biol., Vol. **20**(1), (1975), pp. 1~29.
8) R. Birch and M. Marshall, "Computation of Bremsstrahlung X-ray Spectra and Comparison with Spectra Measured with a Ge(Li) Detector", Phys. Med. Biol., Vol. **24**(3), (1979), pp. 505-517.

9) R.L. Morin and D.E. Raeside, "Bremsstrahlung Spectra Sampling for Monte Carlo Simulations", Phys. Med. Biol., Vol.**27**(2), (1982), pp. 223-228.

10) J.W. Motz, C.E. Dick, A.C. Lucas, R.C. Placious, and J.H. Sparrow, "Production of High-Intensity K X-ray Beams", J. Appl. phys., Vol. **42**, (1971), pp. 2131-2133.

11) J.H. Sparrow and C.E. Dick, "The Development and Application of Monoenergetic X-ray Sources", NBS SP **456**, (1976), pp. 41~45.

12) J.L. Chartier, D. Roman, and A. Bazoge, "Problems of Exposure Measurement with Extended Sources; Case of X-Ray Fluorescence", Nucl. Inst. Meth., Vol. **119**, (1974), pp. 427-444.

13) R.O. Müller, "Spectrochemical Analysis by X-Ray Fluorescence", Plenum press, New York, (1972).

14) N.A. Dyson, "X-Rays in Atomic and Nuclear Physics", Longman, London, (1973).

15) F.H. Attix, W.C. Roesch, and E. Tochilin, "Radiation Dosimetry" Vol. Ⅲ, 2nd. ed., Academic press, London, (1969).

16) G.F. Knoll, "Radiation Detection and Measurement", John Wiley & Sons, Inc., New York, (1979).

17) "Calibration of Dose Meters Used in Radiotherapy", Tech. Rep. Ser. No. **185**, IAEA, (1979).

18) E.P. Bertin, "Principles and Practice of X-ray Spectrometric Analysis", 2nd. ed., Plenum Press, New York, (1975).

19) J.V. Boggs, "A Portable Low-Energy Photon Calibration Source", UCRL-72737 Rev. 1, (1971).

20) T.H. Rogers, "Production of Monochromatic X-Radiation for Microradiography by Excitation of Fluorescent Characteristic Radiation", J. Appl. Phys., Vol. **23**(8), (1952), pp. 881-887.

21) L.S. Birks, R.E. Seebold, A.P. Batt, and J.S. Grosso, "Excitation of Characteristic X-Rays by Protons, Electrons, and Primary X-Rays", J. Appl. Phys., Vol. **3519**, (1964), pp. 2578-2581.

22) J.H. Hubble, "Photon Mass Attenuation and Energy-absorption Coefficients from 1 KeV to 20 MeV", Int. J. Appl. Radiat. Isot., Vol. **33**, (1982), pp. 1269-1290.

23) S.T. Hwang, S.H. Hah, and N.W Park, "Measurement and Analysis of X-ray Spectrum Using Hyperpure Ge Detector", KSRI-MO-25, (1983).

형광 X선을 이용한 100 KeV 이하의 에너지 영역에서의 단색 Photon 선원개발에 관한 연구

한국과학기술원

이 연 명 · 이 건 재

한국표준연구소

하석호 · 황선태 · 이경주

요 약

고순도 물질의 K_α형광 X-선을 이용하여 100 keV 이하에서의 단색 Photon 선원을 개발하였다. 단색 Photon은 방사선 측정기기의 교정에 매우 유용한 것으로 얇은 금속박막의 Radiator에 제동복사선을 조사하여 얻을 수 있다. 본 실험에서는 $_{47}$Ag, $_{50}$Sn, $_{68}$Er, $_{70}$Yb, $_{82}$Pb, 등의 Radiator를 사용하여 20 keV에서 80 keV의 범위에서 단색 Photon을 얻어내어 고순도 HpGe LEPS로 0.64-0.94에 이르는 단색도를 얻어내었고 600cc 얇은창이온전리함을 사용하여 8.3mR/h에서 232.5mR/h에 이르는 선량률을 얻어 내었다.