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Mass Attenuation Coefficients and Related Parameters of Citrine at Different Gamma Rays Energy using Compton Scattering Technique

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Abstract

Citrine is a transparent gemstone, yellow microcrystalline quartz variety. Natural citrine is very rare and will actually lose its color upon heating to more than 200°C, and the yellow color will occur again when the crystal is irradiated with X- or gamma-rays, indicating that attenuation parameters is important to enhancements by irradiation. In this work, the mass attenuation coefficients of citrine were measured at the different energy of gamma-rays using the Compton scattering technique. The results show that, the experimental values of mass attenuation coefficient are in good agreement with the theoretical values. The mass attenuation coefficients increased with the decreasing of the gamma ray energies. This may be attributed to the higher photon interaction probability of citrine at lower energy. This result is a first report of mass attenuation coefficient of citrine at different gamma ray energies.

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Keywords: Citrine; Mass Attenuation Coefficient; Effective Atomic Number; Compton

1. Introduction

Irradiation affects only a gemstone's color. Subatomic particles, such as gamma rays, are used to bombard the electrons of the gem, causing them to be 'knocked' lose, and captured by other atoms. The light-absorbing pattern is thereby changed and as a result, so is the color. In the case of gamma rays, no leftover radioactivity remains.

Citrine is yellow to brownish quartz (silicon dioxide) and resembles yellow topaz. It is colored by hydrous iron oxide, and is found in the same hexagonal crystals as the other varieties of crystalline quartz. Natural citrine

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is much less common than amethyst or smoky quartz, both of which can be heat-treated to turn their color into that of citrine. Most citrine that available is in fact heat-treated amethyst and smoky quartz. This citrine will actually lose its color upon heating to more than 200°C, and the yellow color will occur again when the crystal is irradiated with X- or gamma-rays [1].

Elemental compositions of citrine have been determined by energy dispersive x-ray fluorescence (EDXRF) spectroscopy technique. The advent of commercially available energy-dispersive spectrometer for X-ray fluorescence (EDXRF) measurements has provided an economical and powerful tool for environmental, clinical, chemical, geological and industrial analysis. EDXRF is a non-destructive, fast multi-element technique for analyzing the surface layer and determining major, minor or trace elements in thin and thick samples of all sizes and forms [2].

The knowledge of mass attenuation coefficients and effective atomic number of gemstones are important parameters to enhancements by irradiation. Several authors reported mass attenuation coefficients and effective atomic number of some gemstones as following: Gerward et al. [3,4] introduced the WinXCom program for calculating the mass attenuation coefficients of elements, compounds and mixtures materials. Korkut et al. [2] studied the mass attenuation coefficients of amethyst ore at different gamma ray energies. Midgley [5] measured the X-ray linear attenuation coefficients of sapphire, beryl and almandine garnet at energies 32-66 and 140 keV. Ryzhikov [6] reported the effective atomic number of quartz glass.

In this work, the compositions of citrine have been analyzed by energy dispersive x-rays fluorescence spectrometer (EDXRF). The mass attenuation coefficients and the effective atomic numbers of citrine were measured at different photon energy. The incident photon energy has been changed by Compton scattering technique.

2. Theoretical Backgrounds

2.1 Compton scattering

The inelastic scattering of X-rays and gamma rays from electrons had been known for a decade when the American researcher A.H. Compton showed the relationship between incident and scattered gamma ray energies to be [7]

$$E_{\gamma'} = \frac{E_\gamma}{1+(1-\cos\theta)E_\gamma / mc^2} \qquad (1)$$

where $E_{\gamma'}$ is the scattered gamma rays energy, E_γ is the incident gamma rays energy, θ is the scattering angle, and m is the electron rest mass. This formula is easily derived by assuming a relativistic collision between the gamma ray and an electron initially at rest. Of course, under normal circumstances, all the electrons in a medium are not free but bound. If the energy of the photon, however, is of the order of keV or more, while the binding energy of the electron is of the order of eV, the electron may be considered at rest. The collision is inelastic in the sense that one photon is absorbed and another of different frequency and momentum is emitted.

2.2 Mass attenuation coefficient and effective atomic number

The mass attenuation coefficient is written as [8]

$$\mu_{\mathrm{m}} = \frac{\ln(\mathrm{I_0}/\mathrm{I})}{\rho \mathrm{t}} \qquad (2)$$

Where ρ is the density of material (g/cm^3), I_0 and I are the incident and transmitted intensities and t is the thickness of absorber (cm).

Theoretical values of the mass attenuation coefficients of mixture or compound have been calculated by WinXCom, base on mixture rule [4].

$$\mu_m = \sum_i w_i (\mu_m)_i \tag{3}$$

Where w_i is weight fraction of element in citrine, $(\mu_m)_i$ is mass attenuation coefficient for individual element in citrine. The value of mass attenuation coefficients can be used to determine the total atomic cross-section ($\sigma_{t,a}$) by the following relation [8]

$$\sigma_{t,a} = \frac{(\mu_m)_{alloy}}{N_A \sum_i^n (w_i / A_i)} \tag{4}$$

Where N_A is Avogradro's number, A_i is atomic weight of constituent element of citrine. Also the total electronic cross-section ($\sigma_{t,el}$) for the element is expressed by the following formula [8]

$$\sigma_{t,el} = \frac{1}{N_A} \sum_i^n \frac{f_i A_i}{Z_i} (\mu_m)_i \tag{5}$$

Where f_i is the number of atoms of element i relative to the total number of atoms of all elements in citrine, Z_i is the atomic number of the ith element in citrine. Total atomic cross-section and total electronic cross-section are related to effective atomic number (Z_{eff}) of the compound through the formula [8]

$$Z_{eff} = \frac{\sigma_{t,a}}{\sigma_{t,el}} \tag{6}$$

3. Experimental setup

The compositions of citrine were analyzed by energy dispersive x-rays fluorescence spectrometer (Panalytical Minipal-4). The density of the sample at room temperature is measured by the Archimedes's principle using a sensitive microbalance with xylene as the immersion liquid. The density is calculated according to the formula,

$$\rho = \frac{W_a}{W_a - W_b} \times \rho_b \tag{7}$$

where W_a and W_b is the weight of sample in air and xylene, respectively, and ρ_b is the density of xylene (ρ_b = 0.863 g/cm^3). All weight measurements were used a sensitive microbalance.

The experimental arrangement is shown in Fig. 1. The source system was mounted on a composite of adjustable stands. This setup can move in the transverse direction for proper beam alignment. The ^{137}Cs radioactive source of 15 mCi (555 MBq) strength was obtained from the Office of Atom for Peace (OAP), Thailand. The aluminium rod was used the scattering rod. The Compton scattered γ-rays were measured on a rotatable scintillator detector in the scattering plane by using the 2″×2″ NaI(Tl) detector having an energy resolution of 8% at 662 keV (BICRON model 2M2/2), with CANBERRA photomultiplier tube base model 802-5. The optimum distance between the source and the scatterer was chosen to be 20 cm and that between the scatterer and detector, 20 cm. The spectra were recorded using a CANBERRA PC-based multi-channel analyzer (MCA).

The spectrum on the MCA of detector gave instance counts in each of 1024 bins divided by voltage. To measure the angular dependence of Compton scattering, we first perform a calibration relating the channel number of the MCA spectrum to the energy of known gamma-ray sources. We vary the angle of the scatter detector and acquire measurements on the MCA. The different angles (θ) were used to produce the different gamma rays energies.

An optimum sample thickness ($0.5 \le \mu x \le 5.0$) was selected in this experiment on the basis of the Nordfors criteria [9]. To measure mass attenuation coefficient, we placed the sample between the scattering rod and detector, and detection the acquired MCA spectra of the scattered gamma rays photopeak through sample thickness at different angles. Integrated count rates were determined from Gaussian fits and used to determine an attenuation coefficient.

The statistical error in this experiment calculated from the standard error of 3 items (i) ray-sum measurement, which calculated from experiment, the ray-sum is product of linear attenuation coefficient (μ) with thickness (x), (ii) density measurement and (iii) thickness measurement [8]. Finally, the total standard error has been determined by combining errors for the ray-sum measurement, density measurement and thickness measurement in quadrature.

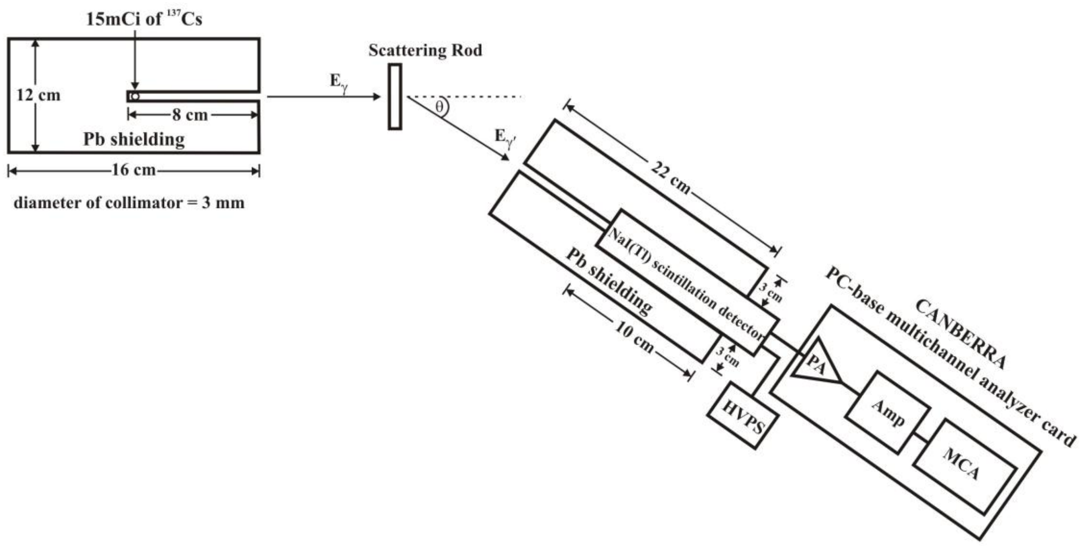

Fig. 1. Schematic of the Compton scattering experiment

4. Validation of Compton scattering system setup

Gamma ray spectrometer is calibrated by using standard calibration sources of different energies (^{241}Am 59.5 keV, ^{133}Ba 356 keV, ^{22}Na 511 keV and ^{137}Cs 662 keV). Each of the calibration sources is placed at the scatterer position and its spectrum is recorded. The gamma rays energy spectrum for the NaI(Tl) detector was a Gaussian shaped peak. For each full energy peak, the centroid and full width at half maximum (FWHM) of the full energy peak were obtained from Gaussian fitting software of Canberra MCA.

The statistical error in this experiment calculated from full width at half maximum (FWHM) of the full energy peak. The width of a Gaussian distribution is related to the standard deviation σ by [7]

$$FWHM = 2\sqrt{2\ln 2}\sigma \qquad (8)$$

Fig. 2 shows the plot of FWHM as function of incident photon energy. The solid curve gives a best fit through the points corresponding to observed FWHM values from standard sources. The FWHM of the Compton photopeaks higher than FWHM of point sources attributed to Compton scattering experiment several broadening effects contributing to our error, and some negligible effects relevant to higher resolution measurements. Inherent to our experimental setup is the spread of the beam profile ($\Delta\theta \approx 3^o$) of the ^{137}Cs source over the surface of the scatter detector that contributes to a Gaussian spread of measured scattering events. The Fermi motion of electrons in the detector has a finite momentum distribution contributing to Lorentzian line broadening beyond the resolution of the NaI scintillator. Additionally, the motion of electrons about the binding potential of the nucleus Doppler broadens the apparent energy of incident photons from our source, thereby broadening the Compton peak for a given scattering angle [10]. The statistical error of each photons scattered in our experiment shown in Table 1.

Table 1 shows the scattered gamma ray energies at different angles. The theoretical values ($E'_{\gamma(th)}$) were calculated by using Eq. (1) and the experimental values ($E'_{\gamma(ex)}$) were measured. The relative difference between theoretical values and experimental values have maximum about 5% at 20^o, this results reflect the good detection system setup of Compton scattering experiment.

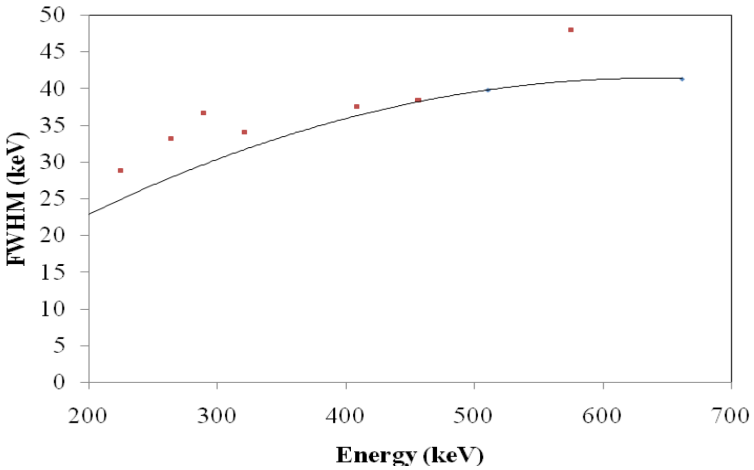

Fig. 2 The full width at half maximum (FWHM) of NaI(Tl) scintillation detector as function of incident photon energy. The solid line is FWHM of point sources and dot is FWHM of Compton energies at different angles

Table 1. The scattered gamma rays energies at different angles

θ (deg)	$E'_{\gamma(th)}$ (keV)	$E'_{\gamma(ex)}$ (keV)	%RD
30	564.09	562.68 ±27.8	0.25
45	479.90	481.59 ±20.7	0.35
60	401.76	398.97 ±16.0	0.69
75	337.72	340.83 ±16.1	0.92
90	288.39	287.28 ±15.6	0.39
105	251.63	252.98 ±14.2	0.54
120	224.92	223.02 ±12.2	0.84

% RD = [(Theoretical value – Experimental value)/ Theoretical value] x 100

5. Results and discussions

The density of citrine is 2.6462 ±0.0082 g/cm^3. The thickness of citrine is 0.415 ±0.001 cm. The chemical compositions of citrine in oxide form are shown in Table 2. It was seen that the major composition of all citrine is SiO_2.

Table. 2 The chemical compositions of citrine in oxide form

Composition	% by weight
SiO_2	83.50
P_2O_5	12.80
CaO	3.40
TiO_2	0.01
CuO	0.06
ZnO	0.03
BaO	0.20

The mass attenuation coefficients of citrine as shown in Table 3 were evaluated from incident (I_0) and transmitted (I) intensities of each energy and compared with theoretical values were calculated by WinXCom program [4]. It has been found that the total mass attenuation coefficients were increased with decreasing of gamma rays energy, which indicates the total interaction is increased. The experimental values of mass attenuation coefficient are in good agreement with the theoretical values as shown in Fig. 3.

The partial interactions of citrine in this energy range as shown in Table 4, were calculated from WinXCom program. It was found that the photoelectric absorption and coherent scattering interactions were increase with decreasing of gamma rays energy. This interaction was found to be small effect on total mass attenuation coefficient in this energy range. The incoherent (Compton) scattering interaction was found to be the main interaction process in this energy range for citrine. It was found to be increased with decreasing of gamma rays energy as shown in Fig. 4.

The effective atomic numbers (Z_{eff}) have been determined using Eq. (6) and are given in Table 3. It was found that the effective atomic numbers is around 10.1 electrons/atom which almost constant in this energy range for citrine. So the effective atomic numbers of citrine not depend on gamma rays energy in this range.

Table 3. The mass attenuation coefficients and effective atomic numbers of citrine at different energies

E_γ'(keV)	$\mu_{m\ (th)}$ ($x10^{-2}$ cm^2/g)	$\mu_{m\ (ex)}$ ($x10^{-2}$ cm^2/g)	%RD	$Z_{eff(th)}$ (e^-/atom)	$Z_{eff(ex)}$ (e^-/atom)
662.00	7.716	7.559 ±0.310	2.03	10.12	9.917 ±0.407
562.68	8.286	8.434 ±0.344	1.79	10.13	10.31 ±0.42
481.59	8.883	8.848 ±0.343	0.39	10.13	10.09 ±0.39
398.97	9.567	9.592 ±0.372	0.26	10.14	10.16 ±0.39
340.83	10.27	10.32 ±0.38	0.49	10.15	10.20 ±0.38
287.28	10.93	10.78 ±0.40	1.37	10.15	10.02 ±0.37
252.98	11.53	11.55 ±0.40	0.17	10.17	10.19 ±0.35
223.02	12.05	12.02 ±0.37	0.25	10.19	10.16 ±0.31

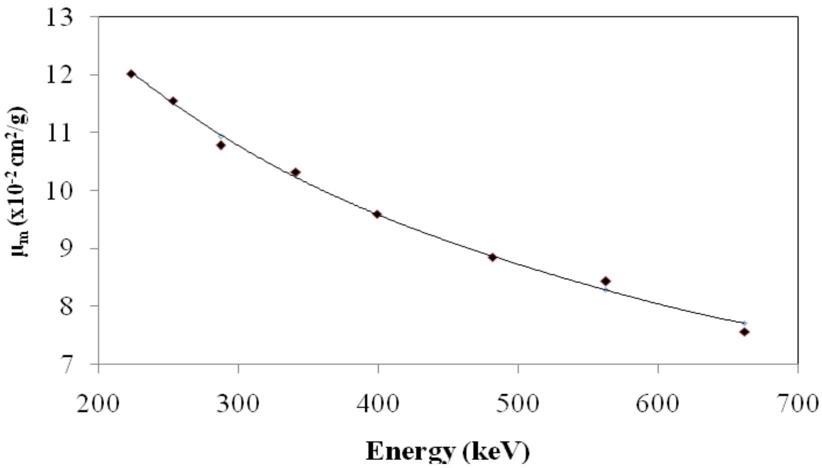

Fig. 3. Variation of mass attenuation coefficient values as a function of gamma rays energies. The line is theoretical value and point in this figure is experimental value

Table 4. Partial interaction of citrine at different gamma rays energies

E'(keV)	Coherent ($x10^{-2}$ cm^2/g)	Incoherent ($x10^{-2}$ cm^2/g)	Photoelectric ($x10^{-2}$ cm^2/g)
662.00	0.033	7.683	0.012
562.68	0.046	8.247	0.018
481.59	0.063	8.802	0.027
398.97	0.091	9.490	0.045
340.83	0.125	10.08	0.070
287.28	0.174	10.73	0.115
252.98	0.224	11.21	0.167
223.02	0.286	11.69	0.245

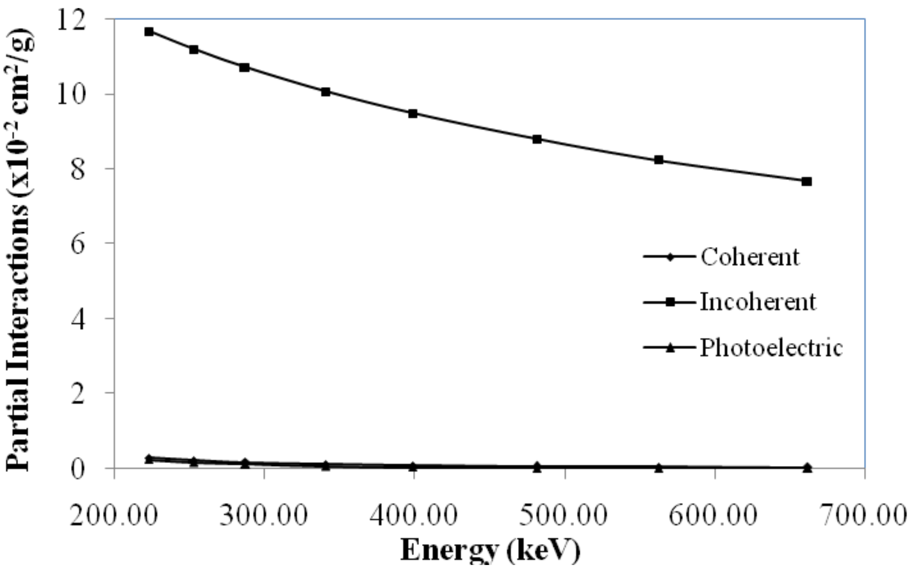

Fig. 4. Variation of partial interactions as a function of gamma rays energies

6. Conclusion

The compositions of citrine were analyzed by energy dispersive x-rays fluorescence spectrometer, the result found that major composition of citrine is SiO_2. The mass attenuation coefficients of citrine were measured at the different energy of gamma rays using the Compton scattering technique. There are corresponding of scattered gamma rays energies between theoretical value and experimental value, reflecting the validation of Compton scattering system setup. The results show that mass attenuation coefficients decrease with the increase in gamma rays energies. The result of partial interaction show the same trend with mass attenuation values which incoherent scattering is main interaction process for citrine in 223-662 keV gamma rays energies. The effective atomic numbers were almost constant, that is the effective atomic numbers of citrine not depend on gamma rays energy in this range.

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