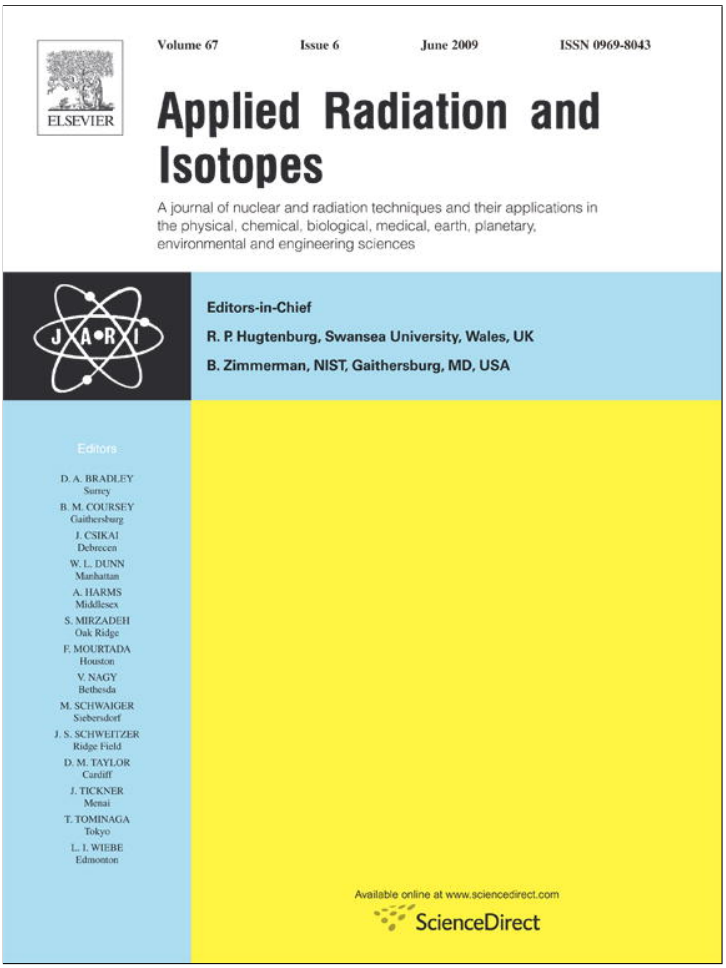

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Using medical accelerators and photon activation to determine Sr/Ca concentration ratios in teeth

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ABSTRACT

This paper describes a photon activation method, studied by using two medical accelerators (energies: 15 and 18 MeV) as photon sources, for determining Sr and Ca levels and Sr/Ca ratios in tooth samples. The radionuclides formed by various photonuclear reactions were measured and identified using a gamma-spectrometry with HPGe detection system. The yields of the corresponding photonuclear reactions and the detection sensitivities for the alkaline earth metals (e.g., Ca, Sr) were surveyed and estimated in relation to the radiation dose. The minimum detectable amount of Sr was estimated to be less than 1 $\mu g\,g^{-1}$, allowing the Sr/Ca ratios in teeth to be determined conveniently. The Sr/Ca ratios in deciduous and permanent tooth samples obtained from local dental clinics were 0.390 and 0.565 $mg\,g^{-1}$, respectively. This photon activation method of determining Sr/Ca ratio in bones and teeth using medical accelerators for cancer treatment is thought to be useful also in biological and archaeological studies.

1. Introduction

The alkaline earth metals calcium, magnesium, strontium, and barium are constituent elements of bones and teeth. The levels of Sr and the derived Sr/Ca ratios are frequently used as indices for understanding prehistoric diet information (Toots and Voorhies, 1965; Balter et al., 2002; Sponheimer et al., 2005), ancient weaning practices (Sillen and Smith, 1984; Mays, 2003), and environmental influences on disease and nutritional status (Brown et al., 2004; Webb et al., 2005). Non-destructive analyses of the major and trace elements present in biological and archaeological samples are of great importance, because these materials are often too precious to be analyzed destructively. Among the non-destructive techniques for elemental analysis, instrumental neutron activation analysis (INAA) is most accurate and used most widely. Nevertheless, the determination of Ca through INAA using the capture reaction of $^{48}Ca(n,\gamma)^{49}Ca$ is not used frequently, because of (1) the low neutron capture cross-section and the low natural abundances of ^{48}Ca, (2) possible presence of limiting radionuclides, such as ^{24}Na, in the counting, and (3) the relatively low counting efficiency of the 3084 keV gamma photons emitted from ^{49}Ca. Moreover, the measurement of the short-lived ($t_{1/2} = 8.72$ min) nuclide ^{49}Ca means that INAA in short-time mode must be used, which requires a fast pneumatic transport system in nuclear facilities. Together, these factors could restrict the detection limits and degrade the resulting precision. The fast neutrons, those from a 14 MeV generator (Hult and Fessler, 1998) are thought to be of limited accessibility and sensitivity. By proton-induced X-ray emission (PIXE), only thin surface portions, such as bone surface and dental enamel, can be analyzed (Anttila and Anttila, 1987; Solis et al., 1996; Lane and Peach, 1997; Williams et al., 2002).

Instrumental photon activation analysis (IPAA) is regarded as being complementary to INAA, and has been used for trace elements in geological, biological, and environmental samples (Sakamoto et al., 1997; Masumoto et al., 1999; Ni et al., 2000; Randa et al., 2003, 2007). Because photonuclear cross-sections are generally smaller than those of thermal neutron capture reactions, a high-intensity photon beam is a prerequisite for IPAA.

At present, very few high energy photon sources are dedicated to IPAA, whereas medical accelerators used for cancer therapy are widely distributed in hospitals; their energies are commonly less than 20 MeV. In this study, we used two medical electron accelerators having energies of 15 and 18 MeV to irradiate tooth

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samples; the activated nuclides emitting characteristic gamma rays were measured to determine the concentrations of several elements of interest. Initially, we surveyed some preferable photonuclear reactions that would be suitable for the analysis of alkaline earth metals and evaluated them in accordance with the respective yields estimated experimentally. Next, we evaluated the detection sensitivities and limits of these elements and compared them based on the bremsstrahlung energies and irradiation and measurement conditions. In this paper we discuss the feasibility of IPAA by the use of accelerators to determine Sr/Ca ratios in bone and tooth samples.

2. Materials and methods

2.1. Sample preparation and irradiation

Powder samples (purity >99%) of $CaCO_3$ (0.3 g), SrO (0.1 g), and BaO (0.1 g) were mixed and pressed into a cylindrical tablet (5 mm$D$ × 5 mm$H$) for use as a reference standard. Two additional pressed standards of MgO (0.5 g) and Na_2CO_3 (0.5 g) were prepared to evaluate the interference with ^{24}Na formed from Mg and Na. To verify chemical purity, these chemicals were digested with HNO_3 and the resulting clear solution were diluted and used for analysis with an inductively coupled plasma atomic emission spectrometer (JY2000, Horiba Jobin Yvon Inc., France). The standard solutions (E. Merck, Darmstadt, Germany) were used for determination of Ca, Sr, etc. The Sr/Ca in the reference standard was determined at 0.353.

The samples of human teeth were collected from local dental clinics (Hsinchu County, Taiwan). All samples were treated with H_2O_2 (10%) for 1 h to remove organic material. The tooth samples weighing ca. 0.2–1.0 g, with the diameter/thickness of less than 5 mm, were wrapped with polyethylene foils. Each set of 1–3 samples and the reference standard were bound tightly and placed at the sample tray (Fig. 1) for photon irradiation.

Two medical accelerators having operating voltages of 15 MV (Primus, Siemens) and 18 MV (Precise, Elekta) were used as irradiation sources. The samples were placed as close as possible to the target (tungsten) in order to receive the maximum photon flux. In the 15 MV accelerator, samples were placed in an acrylic support inserted into the treatment head with a target-to-sample distance of 20 cm (Fig. 1). In the 18 MV accelerator, samples were placed immediately outside the treatment head with a target-to-sample distance of 53 cm. Photoirradiation of samples was conducted at an exposure of 1000 monitor units (MUs) for 20 times within 1 h and opened to a field size of 40 cm × 40 cm in both accelerators, delivering a total radiation dose of 200 Gy at depth of maximum in a water-equivalent phantom with source-surface distance of 100 cm. The actual radiation dose imparted to the samples in each of the two accelerators was measured using a QED photodiode detector (Model 1135, Sun Nuclear Corporation). The photodiode detector having a brass buildup layer of 3.04 g cm^{-2} was suitable for measurement of bremsstrahlung photons of 15–25 MeV emitted from the tungsten targets of the medical accelerators. The energy dependence can be varied by 19% for the QED photodiode for nominal accelerating potential between 1.25 (^{60}Co) and 17 MV (Saini and Zhu, 2007).

In order to realize the photon flux distribution, six pieces of identical gold foils (6 mm$L$ × 6 mm$W$ × 0.05 mm$H$) with an averaged mass of 0.035 ± 0.003 g were evenly placed at the sample tray (Fig. 1) and photoirradiated for 1 h. The induced activities of ^{196}Au ($t_{1/2}$ = 6.18 d), produced through $^{197}Au(\gamma, n)^{196}Au$ reaction, were determined by using the peak at 356 keV and served as a reference to monitor the uniformity of photon flux at the sample position.

2.2. Gamma ray measurement

The irradiated samples and standards were transferred to a low-background lab for counting. The counting system is equipped with a well-type Ge detector (GCW6023, Canberra

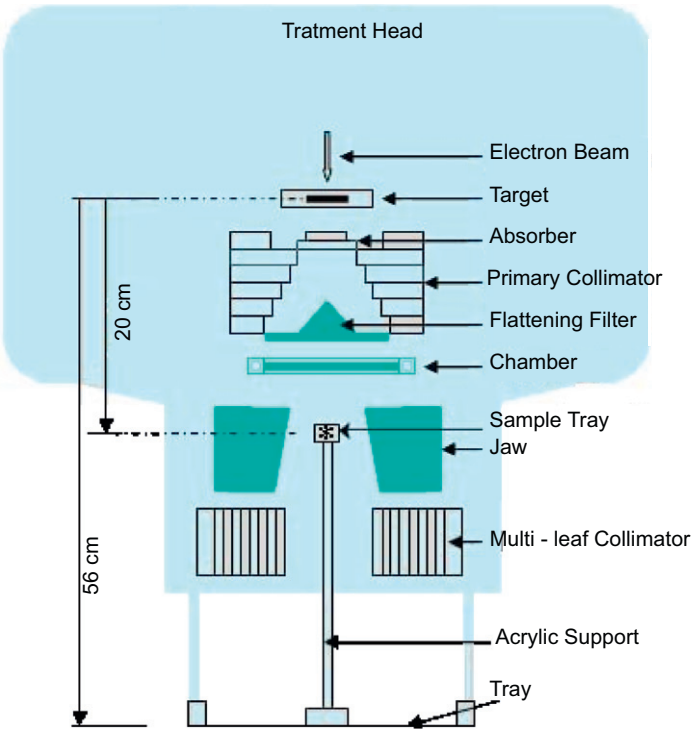

Fig. 1. Schematic representation of the irradiation site (d = 20 cm) within the treatment head of the 15 MV linear accelerator.

Industries, Meriden, CT, USA); the detector has a 60% efficiency and the peak resolution was 2.2 keV (FWHM) at 1332 keV. The measured spectra were collected with a multi-channel analyzer (35-Plus, Canberra Industries, Meriden, CT, USA) and further analyzed using the Sampo-90 gamma-ray spectrum software. For convenience to measure more than one sample, a counting of 3600 s was first conducted within 6 h of irradiation to measure the radionuclides having hour-long half lives (e.g., ^{87m}Sr). Subsequently, 10-hour counting was conducted to determine ^{47}Ca.

2.3. Expression of photonuclear yield

Unlike the neutron capture reaction for INAA, it is difficult to determine the integrated cross-section of a photonuclear reaction experimentally. For medical accelerators, it is more convenient to describe the photon intensity in terms of the dose rate rather than the photon flux. In principle, they are mutually proportional under otherwise the same irradiation conditions. In this study, a dose-based yield (Y_d) is defined as the occurrence possibility of the photonuclear reaction per dose. Table 1 lists the possible photonuclear reactions of alkaline earth metals and their relevant nuclear data.

2.4. Performance evaluation

The performance of IPAA using medical accelerators was evaluated in terms of the detection sensitivity (DS) and the minimum detectable amounts (MDAs) of these elements for the reactions employed. The DS is defined as the counts per mass of a sample; it is obtained by measuring the induced radioactivity under given irradiation and measurement conditions, and is expressed as

$$\mathrm{DS\ (counts/g)} = \frac{A \times I_r \times \in \times t_m \times F_b^{-1} \times e^{-\lambda t_c}}{m} \quad (1)$$

where A is the induced activity (Bq) at end of irradiation; I_r is the intensity of the characteristic gamma rays used for measurement; $\in$ is the detection efficiency of the gamma rays; t_m is the counting time; F_b is the decay correction factor during measurement; t_c is the cooling time; and m is the sample weight. On the other hand, the MDA was calculated based on the Currie's definition (Currie, 1968).

Table 1
Photonuclear reactions of alkaline earth metals and the respective dose-based yields measured in the 15 and 18 MV accelerators.

Element	Photonuclear reactions	Q (MeV)	Y_{15} (Gy^{-1})	Y_{18} (Gy^{-1})	Y_{18}/Y_{15}
Mg	^{25}Mg(γ,p)^{24}Na	12.1	9.07×10^{-20}	1.87×10^{-19}	2.06
Ca	^{43}Ca(γ, p)^{42}K	10.7	NA	NA	NA
	^{44}Ca(γ, p)^{43}K	12.2	NA	NA	NA
	^{48}Ca(γ, n)^{47}Ca	9.94	6.21×10^{-17}	7.36×10^{-17}	1.18
Sr	^{86}Sr(γ, n)^{85m}Sr	11.7	3.18×10^{-17}	7.21×10^{-17}	2.27
	^{86}Sr(γ, n)^{85}Sr	11.5	4.12×10^{-17}	9.64×10^{-17}	2.34
	^{87}Sr(γ, γ′)^{87m}Sr	0.388	NA	NA	NA
	^{88}Sr(γ, n)^{87m}Sr	11.1	4.11×10^{-17}	7.43×10^{-17}	1.81
Ba	^{132}Ba(γ, n)^{131}Ba	9.80	2.59×10^{-16}	5.08×10^{-16}	1.96
	^{134}Ba(γ, n)^{133m}Ba	9.76	3.64×10^{-17}	6.77×10^{-17}	1.86
	^{135}Ba(γ, 2n)^{133m}Ba	16.7	NA	NA	NA
	^{135}Ba(γ, γ′)^{135m}Ba	0.268	NA	NA	NA
	^{136}Ba(γ, n)^{135m}Ba	9.38	6.32×10^{-17}	8.79×10^{-17}	1.39
	^{137}Ba(γ, 2n)^{135m}Ba	16.3	NA	NA	NA

NA: No data available.

2.5. Statistical analysis

All statistical analysis was conducted using the statistical package SPSS 13.0. Continuous variables of Sr/Ca ratio were presented in mean values and standard deviation (SD) and statistically analyzed by Student's t-test.

3. Results and discussion

3.1. Dose-based yield of photonuclear reaction

The dose rates for the 1 h photoirradiation were measured using the photodiode detector; they averaged 0.7794 Gy s^{-1} (source-to-target distance: 20 cm) and 0.1938 Gy s^{-1} (53 cm) in the 15 and 18 MV accelerators, respectively. We used these data to calculate the yields of the photonuclear reactions (Table 1).

Magnesium (^{24}Mg) undergoes a (γ, p) reaction with a relatively low yield, ca. two orders of magnitude lower than those of the other reactions. No measurable end products from the reactions ^{43}Ca(γ, p)^{42}K and ^{44}Ca(γ, p)^{43}K appeared in the spectrum. We identified only the reaction through ^{48}Ca(γ, n)^{47}Ca and used it for the determination of Ca; we suspect that because ^{48}Ca is a neutron-rich nucleus, it undergoes preferably the (γ, n) reaction with a relatively low threshold energy. As a whole, the values of Y_d for the 18 MV bremsstrahlung photons were higher than those for the 15 MV ones, by factors varying from 1.2 to 2.2, depending on the Q values. This factor was increased with the value of Q (Fig. 2), indicating that more photons of sufficiently high energies from the 18 MV accelerator were available for photon activation.

3.2. Estimation of detection sensitivity

We estimated the detection sensitivities of Mg, Ca, Sr, and Ba for given irradiation and measurement conditions in the 15 MV accelerator (Table 2). We also calculated the corresponding MDAs for comparison. Although the ^{48}Ca(γ, n)^{47}Ca and ^{88}Sr(γ, n)^{87m}Sr reactions were of comparable yields, the DS was remarkably higher for Sr than that for Ca due to the higher abundance of ^{88}Sr and higher detection efficiency at 388 keV for ^{87m}Sr (Table 2). In addition, the photo-excitation reaction ^{87}Sr(γ, γ′)^{87m}Sr produces identical radionuclide (Veres, 1980) and contributes to the total production of ^{87m}Sr. This photon excitation reaction should also contribute ^{87m}Sr through ^{87}Sr(γ, γ′)^{87m}Sr reaction. Because its

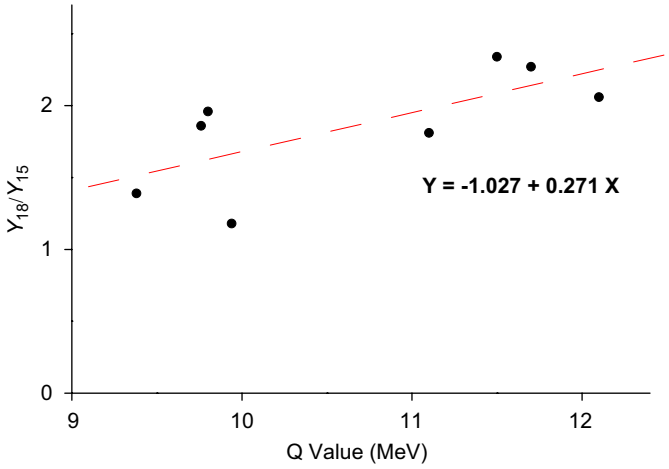

Fig. 2. Yield ratios (Y_{18}/Y_{15}) of the 18–15 MeV photons plotted against the reaction energy (Q value) of the photonuclear reactions. The energy indicated with * in Table 2 were used for evaluation.

Table 2
Detection sensitivities of the alkaline earth metals, obtained using the 15 MV accelerator.

Element	Photonuclear reaction	Abundance (%)	Half life ($t_{1/2}$)	E_γ(I) (keV)	DS (c/g)	MDA (g)
Mg	^{25}Mg(γ, p)^{24}Na	10.0	15.02 h	1369* (100%); 2754 (100%)	1.2×10^{3}	0.012
Ca	^{43}Ca(γ, p)^{42}K	0.135	12.36 h	1525 (18.3%)	NA	NA
	^{44}Ca(γ, p)^{43}K	2.09	22.3 h	373 (87.9%); 397 (10.96%); 618 (78.3%)	NA	NA
	^{48}Ca(γ, n)^{47}Ca	0.187	4.54 days	808 (6.89%); 1297* (74.9%)	1.20×10^{4}	4.16×10^{-3}
Sr	^{86}Sr(γ, n)^{85m}Sr	9.8	67.66 m	151 (12.8%); 232* (84.4%)	2.35×10^{6}	1.33×10^{-5}
	^{86}Sr(γ, n)^{85}Sr	9.8	64 days	514* (99.3%)	6.28×10^{4}	1.62×10^{-3}
	^{88}Sr(γ, n)^{87m}Sr	82.6	2.81 h	388* (81.8%)	4.75×10^{7}	8.88×10^{-7}
Ba	^{132}Ba(γ, n)^{131}Ba	0.101	12.0 days	124 (29.2%); 496* (47.1%)	6.74×10^{3}	0.334
	^{134}Ba(γ, n)^{133m}Ba	2.42	38.9 h	276* (17.5%)	9.66×10^{4}	1.76×10^{-3}
	^{136}Ba(γ, n)^{135m}Ba	7.85	28.7 h	268* (15.6%)	6.45×10^{5}	2.70×10^{-4}

1. NA: No data available.
2. Asterisk (*) indicates the gamma rays used for the determination of DS and MDA.
3. MDA was calculated based on the conditions: photoirradiation with a dose rate of 0.7794 Gy s^{-1} for 1 h, and 10 h counting immediately after irradiation.

contribution was difficult to discriminate, we did not evaluate it in this study. The spectral background at the photopeak of 388 keV was 2×10^{-3} cps; thus the MDA of Sr in a tooth sample (1.0 g) can be calculated to be less than 1 μg g^{-1}. We determined Ca by measuring the activity of ^{47}Ca, which might also be produced through reaction ^{48}Ca(γ, p)^{47}K and its following negatronic decay of ^{47}K ($t_{1/2}=17.5$ s), though the predicted yield is lower. The lowest DS was observed for Mg, presumably because light elements theoretically require relatively high reaction energies and give low yields for photonuclear reactions (Lutz, 1971). Although Ca is less sensitive than Sr to photon activation, its high concentrations in bones and teeth allow to be detected with enough reliability for the determination of Sr/Ca ratios.

3.3. Sample analysis

Barium was not detectable because its concentration in teeth is less than 20 ppm (Aras et al., 1999; Brown et al., 2004), which is lower than its predicted MDA (Table 2). The mean level of Mg in teeth, reported to be 0.2% by weight, is also lower than the predicted MDA for Mg (1.2%, Table 2) based on 1.0 g tooth sample. Mg can be activated by photons to form ^{24}Na, but ^{24}Na can also be generated through secondary neutrons produced mainly from target (tungsten) and surrounding materials by means reaction ^{23}Na(n, γ)^{24}Na. The thermal neutron flux was estimated previously to be 10^{4} cm^{-2} s^{-1} (Chao et al., 2007) around a 15 MV accelerator. Under the irradiation condition we employed in this study, we expected that a tooth sample (1.0 g) containing average concentrations of Mg and Na of 0.2 and 0.4% wt, respectively (Brudevold et al., 1975; Anttila and Anttila, 1987), would generate ca. 4.4×10^{-2} and 2.4 Bq ^{24}Na, respectively. Therefore, the abundance of the end product ^{24}Na did not properly reflect the Mg levels in the tooth samples.

Calcium is the most abundant constituent element in bones and teeth; it is present mainly as calcium phosphate [$Ca_{10}(PO_4)_6(OH)_2$] in the mineral phase (Klepinger, 1984). Table 3 lists the concentrations of Ca and Sr that we determined in teeth collected from local dental clinics; the Sr/Ca ratios are provided for reference. The teeth were divided into two groups: deciduous and permanent. The average Sr/Ca ratio for the deciduous teeth was lower than that for the permanent ones (mean ± SD 0.390 ± 0.110 mg g^{-1} vs. 0.565 ± 0.219 mg g^{-1}, $p=0.033$), even though both sets had comparable calcium contents. In addition, Fig. 3 suggests that the Sr/Ca ratios in the teeth were dependent on age. It is noteworthy that long-term accumulation of Sr in mineralized bone might be responsible for the increase in Sr/Ca ratio in permanent teeth over time (Kawamura et al., 1986; Tanaka et al., 1981).

Table 3
Concentrations of Ca and Sr, and derived Sr/Ca ratios, in human teeth.

Sample	Age (y)	Elemental concentration		Sr/Ca (mg g^{-1})
		Ca (%)	Sr (μg g^{-1})	
Deciduous teeth				
D-1	7	20.1(14)	81(8)	0.402(49)
D-2	11	25.4(13)	83(4)	0.326(24)
D-3	10	26.1(12)	132(5)	0.508(29)
D-4	10	27.7(12)	107(5)	0.385(25)
D-5	10	24.9(11)	131(5)	0.528(31)
D-6	5	26.8(13)	70(3)	0.260(18)
D-7	6	29.1(16)	62(8)	0.213(30)
D-8	13	32.8(15)	177(6)	0.540(30)
D-9	7	33.8(17)	123(6)	0.364(26)
D-10	8	30.9(14)	116(8)	0.375(31)
Mean ± SD				0.390 ± 0.110
Permanent teeth				
P-1	84	21.9(10)	232(7)	1.06(6)
P-2	52	23.9(8)	218(10)	0.912(52)
P-3	57	26.5(9)	93(10)	0.349(41)
P-4	53	21.7(8)	141(6)	0.650(38)
P-5	44	25.4(9)	151(5)	0.594(30)
P-6	35	26.2(14)	109(5)	0.414(30)
P-7	39	28.3(12)	169(7)	0.597(34)
P-8	25	22.7(12)	116(5)	0.513(36)
P-9	39	23.8(10)	100(5)	0.420(28)
P-10	34	33.0(14)	134(5)	0.406(24)
P-11	23	24.2(11)	105(4)	0.434(27)
P-12	29	34.0(15)	147(7)	0.432(29)
Mean ± SD				0.565 ± 0.219
p-value				0.033*

The numbers in parentheses give uncertainty (one standard deviation) of counting statistics in the last digits.
Asterisk (*) indicates Student's test of Sr/Ca ratio between deciduous teeth and permanent teeth is significant at $p<0.05$.

3.4. Other remarks

Both INAA and IPAA are most suitable for analyzing bulk samples of teeth or bones. The advantages of using IPAA medical accelerators rather than INAA for bone analysis are the absence of complicated nuclear reactions, interfering and limiting radionuclides in the gamma ray spectra. A typical gamma ray spectrum was presented in Fig. 4 for a tooth sample. The characteristic

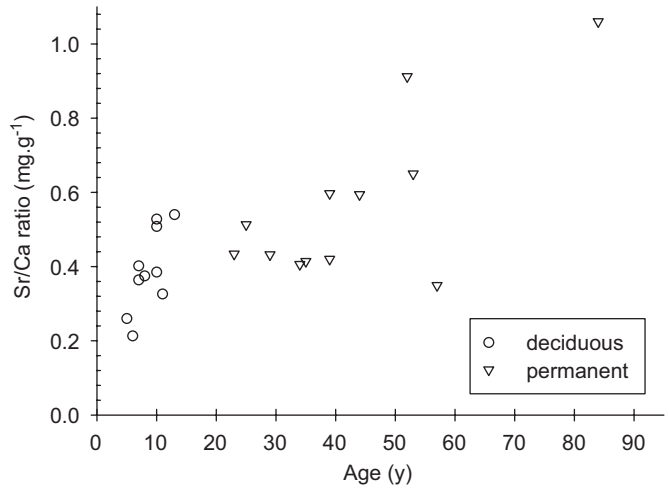

Fig. 3. Age-dependence of Sr/Ca ratios in teeth.

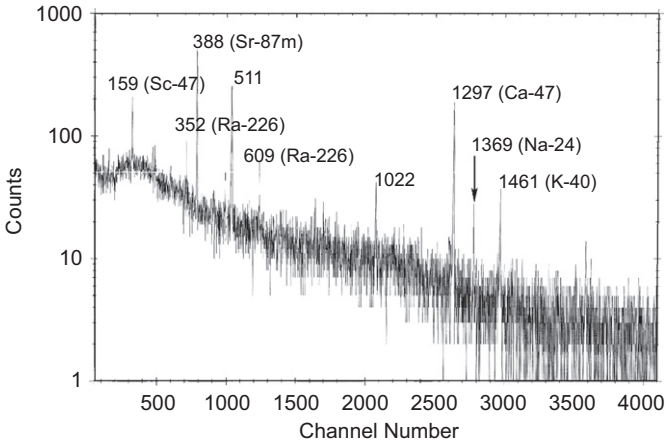

Fig. 4. Gamma ray spectrum of a deciduous tooth irradiated with 15 MeV bremmstrahlung photons. Irradiation time: 1 h (dose rate = 0.7794 Gy s^{-1}); cooling time: 4 h; counting time: 10 h. The peaks at 352, 609 and 1461 keV are from naturally occurring radionuclides ^{226}Ra and ^{40}K, respectively; the annihilation peak (511 keV) and the respective summed peak (1022 keV) are also indicated.

gamma rays at 388 and 1297 keV clearly allow the identification and measurement of Sr and Ca, respectively. The peak at 159 keV in the spectrum arose from ^{47}Sc ($t_{1/2} = 3.42$ days), which was produced through the decay of ^{47}Ca during counting; in contrast, the peak at 1369 keV arose mainly from the neutron activation of Na present in the tooth. Although the concentration of Ca was higher than that of Sr by three or four orders of magnitude, we collected comparable counts for these elements in the measured spectra.

For monitoring the uniformity of photon flux at the sample tray, the ^{196}Au activities of the six gold foils were measured and averaged as 7052 ± 164 Bq/g, or a relative standard deviation of 2.3%, which suggested that the uniformity of photon flux contribute little to the uncertainty in the photon activation experiment.

Attenuation of photons in the tooth sample [$Ca_{10}(PO_4)_6(OH)_2$] and the reference standard can be calculated using the mass attenuation coefficients (Hubbell and Seltzer, 2004). The transmission of 10 MeV photons passing through 1 mm tooth sample and the reference standard are 99.67% and 99.47% respectively, which reduce to 98.37% and 97.27% as the thickness were 5 mm. It implied that the uncertainty due to photon attenuation in the tooth samples (< 1.0 g) in the work is less than 3% and can be negligible for the determination of Sr/Ca ratio.

For an irradiated tooth (1.0 g) containing 25% Ca and 100 ppm Sr, we estimated the induced activities from ^{47}Ca and ^{87m}Sr to be 3 and 5.3 Bq, respectively, at end of the irradiation. These levels were much lower than those obtained using NAA, and are far from the exempt activities for ^{47}Ca (10 Bq/g or 10^6 Bq) and ^{87m}Sr (100 Bq/g or 10^6 Bq) (IAEA, 1996), suggesting that radiation protection practices for handling such samples should not be mandatory.

4. Conclusions

In this study we investigated the potential application of photon activation using medical accelerators for the determination of Sr/Ca ratios in tooth and bone samples. It is indicated that Sr can be determined by IPAA with high sensitivity, thus the concentrations of Ca and Sr in teeth and the derived Sr/Ca ratios, were determined readily. Several other radionuclides, beside ^{87m}Sr and ^{47}Ca, were present in the measured spectra, and the induced activities in the irradiated samples cannot be a concern of radiation safety. This non-destructive technique for the determination of Sr/Ca ratios is perfectly suited to the analysis of precious samples. Presently, the number of research reactors employing INAA is diminishing. At the same time, medical accelerators for cancer treatment are increasingly being installed worldwide; these systems, which can be used as convenient photon sources for IPAA, are much more accessible than nuclear reactors. Our photon activation method using medical accelerators for the determination of Sr/Ca ratios in teeth appears to be practical for biological and archaeological studies; it could be used as an alternative, or a complementary technique to other currently employed non-destructive methods.

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