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The Yields of Photonuclear Reactions in Titanium and Vanadium with Bremsstrahlung up to 60 MeV

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The yields of a number of radioactive nuclei produced by (γ,xnyp) reactions on titanium and vanadium targets have been determined at four different bremsstrahlung maximum energies, 30, 45, 55 and 60 MeV, with the aid of radiochemical methods and gamma-ray spectrometric measurements. The linear electron accelerator of Tohoku University was the bremsstrahlung source. Yields have been expressed as production rates of given radioactive end-products relative to that of the $^{12}C(\gamma,n)^{11}C$ process. Over the range of energies investigated, the yield of each nuclide increases with increasing maximum energy. The yield of a reaction with the emission of more than one nucleon appeared to be strongly energy dependent. Reaction paths leading to the formation of each nuclide were investigated. The photoproduction of ^{47}Ca from titanium has been observed.

In previous work,[1-6] a number of problems involving reaction yields and applicabilities of induced activities to the activation analysis of many elements have been investigated in the energy region of 20 MeV bremsstrahlung. This energy corresponds to giant resonance for photon absorption and, in this energy region, the main nuclear events are the emission of one neutron, one proton, or occasionally one alpha-particle. The excitation of a nucleus by gamma-rays with higher energies gives rise to more complex events. Apart from reactions with the emission of a single particle, those involving the emission of just two or more nucleons become important. Studies of production rates or yields of such photonuclear reactions are helpful in understanding the processes by which particle emissions take place, as well as in considering the applicability of the resultant activities to activation analysis.

Titanium and vanadium, on the other hand, when irradiated with high-energy photons, undergo photodisintegrations leading to the production of ^{45}Ti, ^{48}V, $^{46-48}Sc$ and other nuclides, which are convenient for the gamma-ray spectrometric determinations of these elements. Abe[7] reported methods for the determination of titanium in iron and vanadium, utilizing the 20 MeV linear electron accelerator. Since yield is a strong function of photon energy, it should be noted that higher sensitivities are expected with an accelerator capable of producing bremsstrahlung radiation of higher energies.

Although a number of studies have been performed on photonuclear reactions in the region of medium weight nuclei,[8-12] relatively little has been determined with regard to photonuclear properties of titanium and vanadium at energies beyond giant resonance.

Sherwood *et al.*[13] determined the cross-sections, integrated to 31 MeV, of photonuclear reactions involving titanium nuclides, and Pai *et al.*[14] measured the yields of protons ejected from titanium nuclides irradiated by 22 and 30 MeV bremsstrahlung.

The present report describes some experimental details on the relative yields of various nuclides

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resulting from the irradiation of titanium and vanadium targets by bremsstrahlung with energies up to 60 MeV. The linear electron accelerator of Tohoku University was used as the bremsstrahlung source, and yields were measured for each nuclide at just four different maximum energies, 30, 45, 55 and 60 MeV, with the aid of radiochemical methods.

Experimental

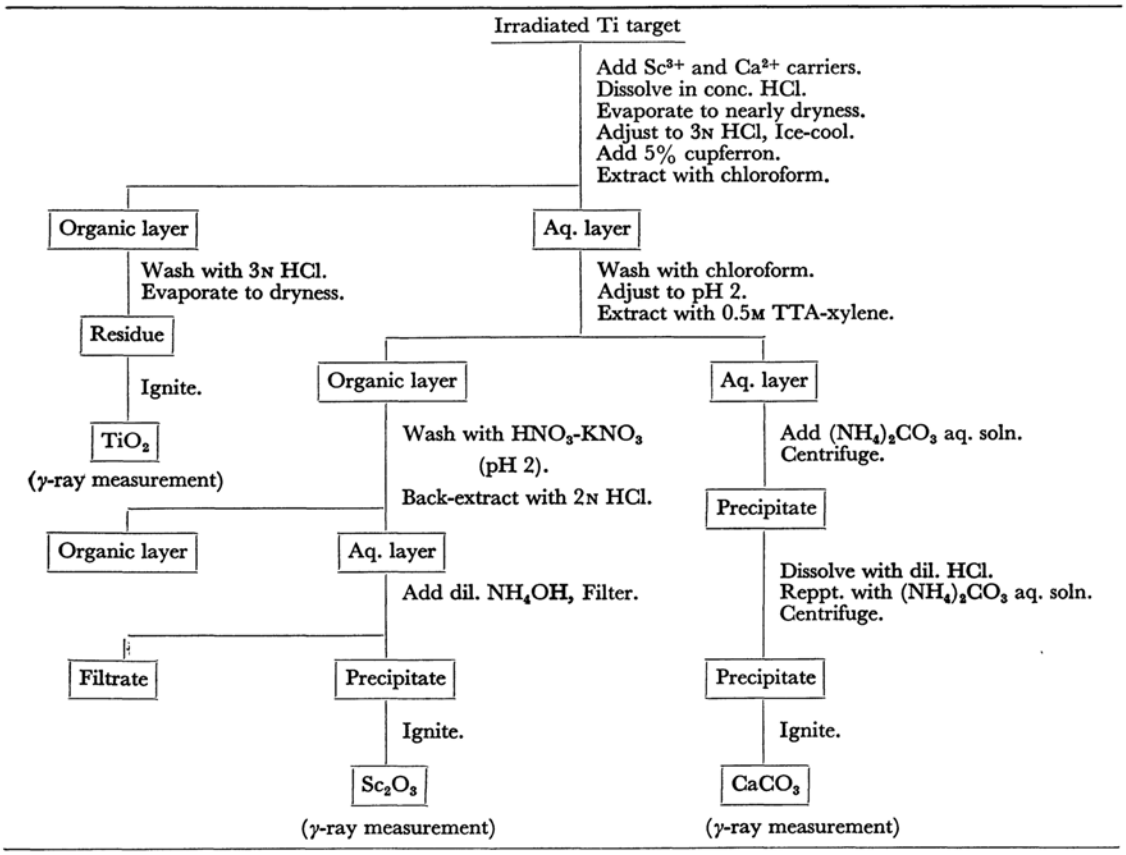
Target Materials. The target materials used were titanium of 99.9% purity, in the form of the elemental powder, and "specpure" grade vanadium pentoxide. A definite amount of each sample (approximately 100 mg) was wrapped in thin aluminum foil and made into a small disk with a diameter of 7 mm and a thickness of 3 mm.

Linac and Irradiation. The 300 MeV linear electron accelerator of Tohoku University was the bremsstrahlung source. The machine was operated with the "High Current" accelerating section which provided a high intensity electron current, the peak current being at least 100 mA with energies up to 100 MeV.[15] The beam was deflected 90° from the original beam path of the accelerating section, and again deflected 45° onto the beam exit window. The spread of electron energies in this operating condition is 3% from the selected value. The electron beam produced bremsstrahlung in a platinum converter with a thickness of 3 mm located 3 cm from the beam exit window. In the energy range from 30 to 60 MeV, used in these experiments, the average beam current was at least 30μA, measured at the position of the converter using a current monitor. Targets were set in a water-cooled target holder on the bremsstrahlung beam axis immediately behind the converter. A typical irradiation was terminated in 1 to 2 hr. The bremsstrahlung flux was monitored using a thin sheet of gold (10μ thick, 7 mmφ) adjacent to the samples.

Radiochemical Separations. After irradiation, the target materials were dissolved along with the addition of scandium and calcium carriers, and subjected to separation procedures diagrammed in Tables 1 and 2. The final precipitates were packed in a small glass tube having an internal diameter of 4 mm for the determination of chemical yield and for gmama-ray counting.

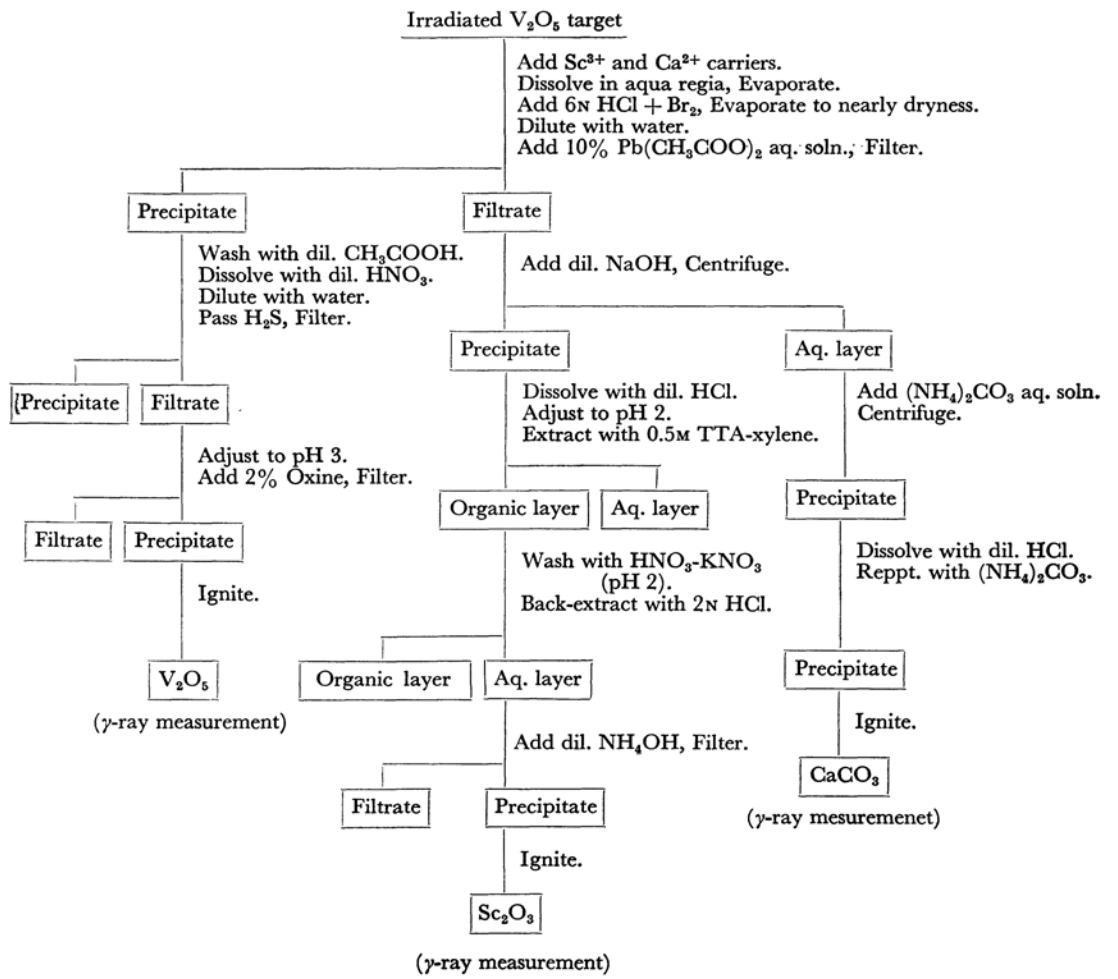
Radioactivity Measurement. For measurements of gamma-ray spectra, a 3″ dia. × 3″ NaI(Tl) detector coupled to a 800-channel pulse-height analyzer made

TABLE 1. SEPARATION SCHEME FOR TITANIUM, SCANDIUM AND CALCIUM ACTIVITIES FROM IRRADIATED TITANIUM TARGET



15) In many cases, the pulse repetition rate was 200 pps with a pulse width of 3μ sec.

TABLE 2. SEPARATION SCHEME FOR VANADIUM, SCANDIUM AND CALCIUM ACTIVITIES FROM IRRADIATED VANADIUM TARGET



by Tokyo Shibaura Electric Co. Ltd., was employed. Counting efficiencies had been previously determined as a function of source distance and gamma-ray energy. The sample to be measured was centered in a Lucite cylinder with walls thick enough to absorb positions from the decay of any positron emitters. Decay curve analyses were performed to distinguish and to separate the undesired activities. The gamma-ray energies used in the yield determination are given in Table 3.

Yield Determination. Yield is defined as the production rate of a given radioactive end-product per atomic weight of the target element when a standard amount of bremsstrahlung radiation with a given maximum energy is passed through the target during the irradiation period. The initial rates of the various activities present were determined from a decay curve analysis of the counting rate of the relevant photopeak area. Corresponding saturation rates were computed and normalized for bremsstrahlung intensity, which was measured by the amount of ^{196}Au activity in a gold foil irradiated together with the target. After correcting for chemical yields, counting efficiencies, branching

TABLE 3. SOME NUCLEAR DATA FOR NUCLIDES USED FOR YIELD DETERMINATION

Nuclide	Half-life	Gamma-ray used	
		Energy (MeV)	Branching ratio (%)
^{45}Ti	3.09 hr	0.511[a)]	168
^{44}Sc	3.92 hr	0.511[a)]	188
^{46}Sc	83.9 d	1.12	100
^{47}Sc	3.43 d	0.160	73
^{48}Sc	1.83 d	1.31	100
^{47}Ca	4.53 d	1.30	76.3
^{48}V	16.0 d	1.31	97

a) Annihilation radiation.

ratio[16] and the internal conversion electrons,[17] data were expressed as yields.

These yields were expressed relative to that of the $^{12}C(\gamma,n)^{11}C$ process, which was measured by the amount of 0.511 MeV gamma-rays arising from the annihilation of positrons in a standard carbon disk[18] irradiated together with the target.

Results and Discussion

Gamma-Ray Spectra. Figures 1—3 show gamma-ray spectra obtained for separated activities obtained from a titanium target irradiated with 60 MeV bremsstrahlung. A typical gamma-ray spectrum of a titanium fraction is shown in Fig. 1. The decay curve of the area under the 0.511 MeV photopeak showed a half-life of 3.09 hr, which agrees with the literature value of ^{45}Ti. The gamma-ray spectra of the scandium fraction separated from an irradiated titanium target show ones composed of ^{44}Sc, ^{46}Sc, ^{47}Sc and ^{48}Sc. The production of ^{47}Ca from a titanium target is revealed by the characteristic gamma-ray photopeaks at 0.49, 0.81 and 1.30 MeV, as shown for a typical gamma-ray spectrum in Fig. 3.[19]

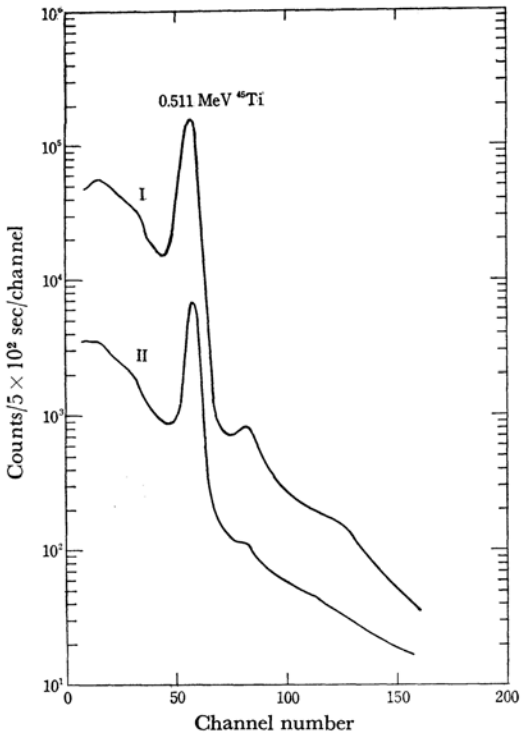


Fig. 1. Gamma-ray spectra of titanium activities.

Irradiation: 60 MeV (45 μA); 1 hr
Source distance:
8 cm from 3″ dia. ×3″ NaI(Tl) crystal
I : 8.8 hr after irradiation
II: 24 hr after irradiation

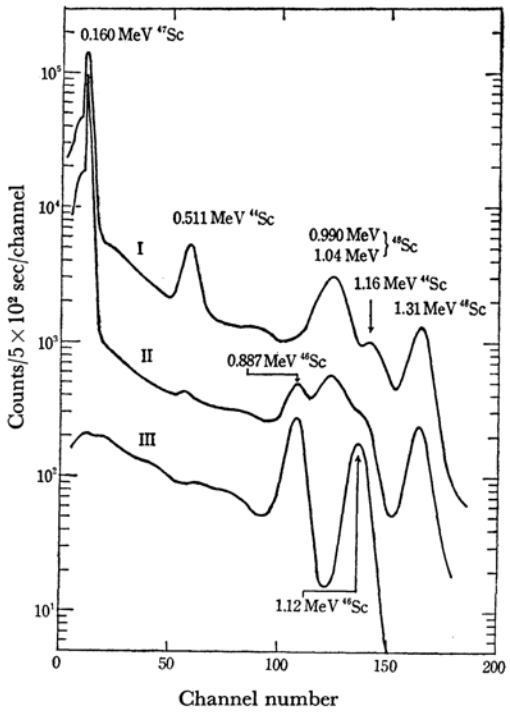


Fig. 2. Gamma-ray spectra of scandium activities separated from titanium target.

Irraidation: 60 MeV (45 μA); 1 hr
Source distance:
8 cm from 3″ dia. ×3″ NaI(Tl) crystal
I : 17.1 hr after irradiation
II : 5.2 d after irradiation
III: 38 d after irradiation

The gamma-ray spectra of the vanadium and scandium fractions from a vanadium target irradiated with 60 MeV bremsstrahlung are shown in Figs. 4 and 5. Photoproductions of vanadium activities other than 16.0-day ^{48}V could not be observed, and no ^{44}Sc activities occurred to any appreciable amount in an irradiated vanadium target under the present experimental conditions. In addition, no gamma-activity could be detected in the calcium fraction.

Reaction Path. The most abundant naturally

16) The decay schemes used were those listed in C. M. Lederer, J. M. Hollander and I. Perlman, "Table of Isotopes," sixth Ed., John Wiley & Sons, Inc., New York (1967).

17) The tables in M. E. Rose, "Internal Conversion Coefficients," North-Holland Publ. Co., Amsterdam (1958), were used.

18) From the carbon rod used as an electrode in emission spectroscopy, small pieces 7 mm $\phi \times 3$ mm were cut out and used.

19) The gamma-ray at 0.160 MeV is due to the growth decay of ^{47}Sc, the daughter nuclide of ^{47}Ca.

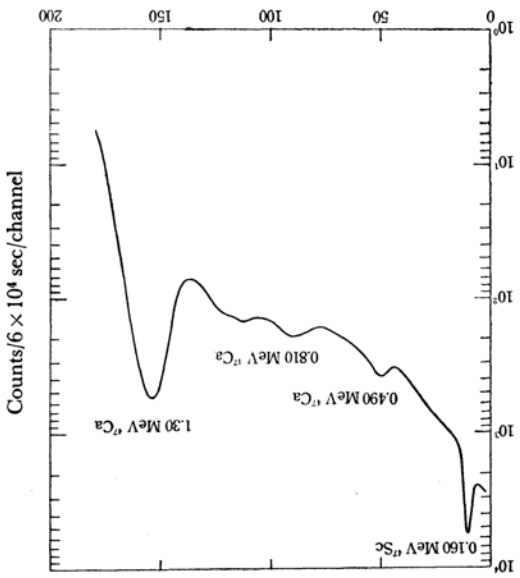


Fig. 3. Gamma-ray spectrum of calcium activities.
Irradiation: 60 MeV (45 μA); 1 hr
Source distance:
5 cm from 3″ dia × 3″ NaI(Tl) crystal
3.0 d after irraditation

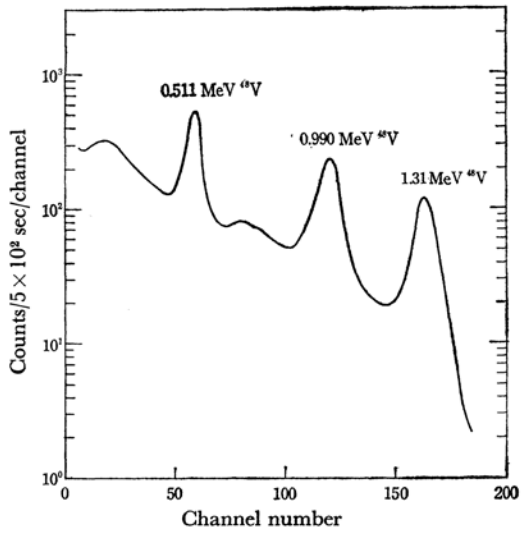


Fig. 4. Gamma-ray spectrum of vanadium activities.
Irradiation: 60 MeV (45 μA); 1 hr
Source distance:
8 cm from 3″ dia × 3″ NaI(Tl) crystal
5.2 d after irradiation.

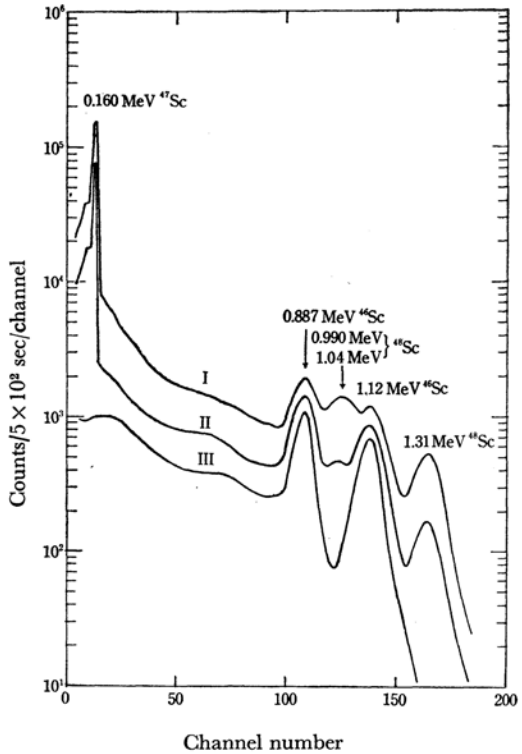


Fig. 5. Gamma-ray spectra of scandium activities separated from vanadium target.
Irradiation: 60 MeV (45 μA); 1 hr
Source distance:
8 cm from 3″ dia. × 3″ NaI(Tl) crystal
I : 2.5 d after irradiation
II : 5.2 d after irradiation
III : 33 d after irradiation

occurring isotope of vanadium is ^{51}V (99.76%). This fact makes the assignments of parent isotopes for observed radionuclides convenient. Titanium, on the other hand, has five naturally occurring isotopes: ^{46}Ti (7.93%), ^{47}Ti (7.28%), ^{48}Ti (73.94%), ^{49}Ti (5.51%) and ^{50}Ti (5.34%). Competitive nuclear reactions should occur in the energy region covered in this experiment. In Tables 4 and 5 are listed the possible nuclear reactions together with their calculated mass thresholds[20] for the production of each radioactive end-product from both elements with natural compositions. In many cases, radionuclides can be formed through several different reaction paths. Photoproduction of ^{45}Ti from vanadium was excluded by considering the natural abundances and threshold energies. Almost all reactions leading to the photoproduction of ^{44}Sc from the parent ^{51}V have threshold energies greater than 50 MeV. This fact makes unlikely a detectable contribution from photonuclear reactions in vanadium to scandium gamma-ray spectra. Finally, the production of ^{47}Ca from vanadium is also unlikely. The reactions producing ^{47}Ca are: ^{50}V $(\gamma,3p)^{47}$Ca, $-Q=28.71$ MeV; ^{51}V$(\gamma,3pn)^{47}$Ca, $-Q=39.75$ MeV; ^{51}V$(\gamma,2pd)^{47}$Ca, $-Q=37.52$ MeV; and ^{51}V$(\gamma,^3Hep)^{47}$Ca, $-Q=32.03$ MeV. Restric-

20) Nuclidic mass values used were those listed in G. Friedlander, J. W. Kennedy and J. M. Miller, "Nuclear and Radiochemistry," 2nd Ed., John Wiley & Sons Inc., New York (1964), p. 533.

TABLE 4. CALCULATED MASS THRESHOLDS FOR PHOTONUCLEAR REACTIONS ON TITANIUM NUCLEI

Reaction	$E_{th}(-Q)$ (MeV)	Reaction	$E_{th}(-Q)$ (MeV)
^{46}Ti(γ, n)^{45}Ti	13.19	^{47}Ti(γ, p)^{46}Sc	10.45
^{47}Ti(γ, 2n)^{45}Ti	22.07	^{48}Ti(γ, pn)^{46}Sc	22.07
^{48}Ti(γ, 3n)^{45}Ti	33.70	^{48}Ti(γ, d)^{46}Sc	19.86
^{49}Ti(γ, 4n)^{45}Ti	42.77	^{49}Ti(γ, p2n)^{46}Sc	30.23
^{50}Ti(γ, 5n)^{45}Ti	52.78	^{49}Ti(γ, dn)^{46}Sc	28.08
		^{49}Ti(γ, t)^{46}Sc	21.75
^{46}Ti(γ, pn)^{44}Sc	21.67	^{50}Ti(γ, p3n)^{46}Sc	41.31
^{46}Ti(γ, d)^{44}Sc	19.44	^{50}Ti(γ, d2n)^{46}Sc	38.95
^{47}Ti(γ, p2n)^{44}Sc	30.56	^{50}Ti(γ, tn)^{46}Sc	32.70
^{47}Ti(γ, dn)^{44}Sc	28.33		
^{47}Ti(γ, t)^{44}Sc	22.07	^{48}Ti(γ, p)^{47}Sc	11.44
^{48}Ti(γ, p3n)^{44}Sc	42.18	^{49}Ti(γ, pn)^{47}Sc	20.36
^{48}Ti(γ, d2n)^{44}Sc	39.95	^{49}Ti(γ, d)^{47}Sc	17.35
^{48}Ti(γ, tn)^{44}Sc	33.69	^{50}Ti(γ, p2n)^{47}Sc	30.52
^{49}Ti(γ, p4n)^{44}Sc	50.32	^{50}Ti(γ, dn)^{47}Sc	28.30
^{49}Ti(γ, d3n)^{44}Sc	48.09	^{50}Ti(γ, t)^{47}Sc	22.04
^{49}Ti(γ, t2n)^{44}Sc	41.84		
^{50}Ti(γ, p5n)^{44}Sc	61.27	^{49}Ti(γ, p)^{48}Sc	11.35
^{50}Ti(γ, d4n)^{44}Sc	59.04	^{50}Ti(γ, pn)^{48}Sc	22.29
^{50}Ti(γ, t3n)^{44}Sc	52.79	^{50}Ti(γ, d)^{48}Sc	20.07
		^{49}Ti(γ, 2p)^{47}Ca	20.77
		^{50}Ti(γ, 2pn)^{47}Ca	31.70
		^{50}Ti(γ, pd)^{47}Ca	29.48
		^{50}Ti(γ, ^{3}He)^{47}Ca	23.99

TABLE 5. CALCULATED MASS THRESHOLDS FOR PHOTONUCLEAR REACTIONS ON VANADIUM NUCLEI

Reaction	$E_{th}(-Q)$ (MeV)	Reaction	$E_{th}(-Q)$ (MeV)
^{50}V(γ, 2n)^{48}V	20.88	^{50}V(γ, 2pn)^{47}Sc	27.52
^{51}V(γ, 3n)^{48}V	31.93	^{50}V(γ, pd)^{47}Sc	25.31
		^{50}V(γ, ^{3}He)^{47}Sc	19.81
^{50}V(γ, 2p2n)^{46}Sc	38.17	^{51}V(γ, 2p2n)^{47}Sc	38.56
^{50}V(γ, pdn)^{46}Sc	35.95	^{51}V(γ, pdn)^{47}Sc	36.34
^{50}V(γ, 2d)^{46}Sc	33.72	^{51}V(γ, 2d)^{47}Sc	34.11
^{50}V(γ, ^{3}Hen)^{46}Sc	30.46	^{51}V(γ, ^{3}Hen)^{47}Sc	30.84
^{50}V(γ, pt)^{46}Sc	29.69	^{51}V(γ, pt)^{47}Sc	30.08
^{50}V(γ, α)^{46}Sc	9.88	^{51}V(γ, α)^{47}Sc	10.26
^{51}V(γ, 2p3n)^{46}Sc	49.21		
^{51}V(γ, pd2n)^{46}Sc	46.99	^{50}V(γ,2p)^{48}Sc	19.29
^{51}V(γ, 2dn)^{46}Sc	44.76	^{51}V(γ, 2pn)^{48}Sc	30.33
^{51}V(γ, ^{3}He2n)^{46}Sc	41.50	^{51}V(γ,pd)^{48}Sc	28.10
^{51}V(γ, ptn)^{46}Sc	40.73	^{51}V(γ,^{3}He)^{48}Sc	22.62
^{51}V(γ, dt)^{46}Sc	38.51		
^{51}V(γ, d, n)^{46}Sc	20.92		

tions posed by the high potential barriers are inherent in these reactions.

Reaction Yield. The yields relative to ^{12}C (γ,n)^{11}C obtained in this experiment are given as a function of the bremsstrahlung maximum energy in Fig. 6. The cross-sectional curve for the reaction ^{12}C(γ,n)^{11}C, reported by Cook *et al.*,[21] has a sharply-peaked resonance centered at 23 MeV, and shows a high energy tail extending at least to 60 MeV. The energy range used herein is well beyond the giant resonance of the ^{12}C(γ,n)^{11}C reaction, and the yield curve for this reaction exhibits a slight increase with increasing energy. Over the range of energies investigated, the relative yield of reactions involving the emission of a single particle do not vary significantly. The yield of a reaction with the emission of more than one nucleon seems likely to be strongly energy dependent.

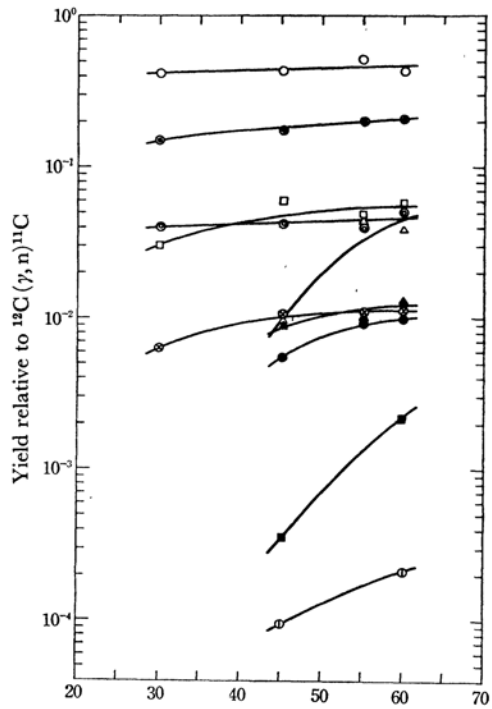


Fig. 6. The yield relative to ^{12}C(γ,n)^{11}C as a function of bremsstrahlung maximum energy.

○ : ^{45}Ti from titanium
● : ^{44}Sc from titanium
◎ : ^{46}Sc from titanium
⊙ : ^{47}Sc from titanium
⊗ : ^{48}Sc from titanium
△ : ^{48}V from vanadium
▲ : ^{46}Sc from vanadium
□ : ^{47}Sc from vanadium
■ : ^{48}Sc from vanadium
◐ : ^{47}Ca from titanium

^{45}Ti from Titanium. The main reaction which can be attributed to the production of ^{45}Ti is the (γ,n) reaction on ^{46}Ti. With high energies, however, contributions from both the ^{47}Ti(γ,2n)^{45}Ti and ^{48}Ti(γ,3n) ^{46}Ti reactions to the total production rate should be taken into account.

$^{44,46-48}$Sc from Titanium. Since ^{44}Sc activity could

21) B. C. Cook, J. E. E. Baglin, J. N. Bradford and J. E. Griffin, *Phys. Rev.*, **143**, 724 (1966).

TABLE 6. THE RELATIVE YIELDS FOR (γ, p) REACTIONS ON TITANIUM NUCLEI EXPRESSED AS THOSE PER MOLE

Reaction	Yield per mole relative to $^{12}C(\gamma, n)^{11}C$			
	30 MeV	45 MeV	55 MeV	60 MeV
$^{47}Ti(\gamma, p)^{46}Sc$	5.4×10^{-1}	5.8×10^{-1}	5.4×10^{-1}	6.3×10^{-1}
$^{48}Ti(\gamma, p)^{47}Sc$	2.0×10^{-1}	2.4×10^{-1}	2.8×10^{-1}	2.8×10^{-1}
$^{49}Ti(\gamma, p)^{48}Sc$	1.2×10^{-1}	2.1×10^{-1}	2.2×10^{-1}	2.1×10^{-1}

not be detected from irradiated titanium in the energy region of 30 MeV bremsstrahlung, its production rate due to the reactions $^{46}Ti(\gamma,d)^{44}Sc$, $^{46}Ti(\gamma,pn)$ ^{44}Sc and $^{47}Ti(\gamma,t)$ ^{44}Sc would be very small. The upward trend of the yield curve for ^{44}Sc with increasing energy is likely attributable to the reactions (γ,tn), (γ,d2n) and (γ,p3n) in ^{48}Ti.

In this experiment, the gamma-activity attributed to ^{44m}Sc is too weak to be clearly distinguished from those due to the decay of ^{44}Sc and other products. The reason responsible for this behavior is the spin difference between the two isomeric states. In many cases in which an excited state of the residual nucleus can decay to either isomer through intermediate states in gamma-cascades, gamma-transitions of low multipolarity are accorded preference over those of high multipolarity.[4] The spins and parities in ^{44}Sc and ^{44m}Sc are 2^+ and 6^+, respectively, and those of the target nuclides are $0^+(^{46}Ti)$, $5/2^-(^{47}Ti)$ and $0^+(^{48}Ti)$. Since the spin in ^{44}Sc is closer to that of the target nuclides than in ^{44m}Sc, the relative probability of forming these two states is expected to be favorable of ^{44}Sc. The main reaction producing ^{46}Sc from titanium may by the (γ,p) reaction on ^{47}Ti. The $^{48}Ti(\gamma,pn)^{46}Sc$ and $^{48}Ti(\gamma,d)^{46}Sc$ reactions would occur by a considerable amount at higher energies. By taking the abundance of ^{48}Ti into account, almost all ^{47}Sc comes from the parent ^{48}Ti through the (γ,p) process, though a considerable amount may be due to the $^{50}Ti(\gamma,pn)^{48}Sc$ reaction.[22]

Assuming that all of the observed ^{46}Sc, ^{47}Sc and ^{48}Sc indeed arise from the parent nuclides through (γ,p) processes it is possible to convert each yield to that per mole of target nuclide, instead of the yield per atomic weight of target element. In Table 6 are given the results thus obtained. The yield values for the production rates of ^{46}Sc, ^{47}Sc and ^{48}Sc decrease in that order.[23] These results are consistent with the fact that the cross-section for photoproton emission from a neutron excess nuclide is expected to be small.

^{47}Ca from Titanium. The formation of ^{47}Ca could not be attributed to a unique reaction, but to the competitive reactions listed in Table 4. The yields amount to about 1—2 per cent of those for the reaction producing ^{48}Sc, which can be taken as equivalent to the emission of one excess proton from a parent nuclide.

^{48}V from Vanadium. An irradiated vanadium target did not show any ^{48}V activity at 30 MeV, but its production rate increases markedly with increasing maximum energy. It seems likely that almost all of the ^{48}V comes from ^{51}V through the (γ,3n) process.

$^{46-48}Sc$ from Vanadium. For the production of ^{47}Sc and ^{46}Sc from ^{51}V, the only paths which have mass thresholds below 30 MeV are the (γ,α) and (γ,αn) reactions. Of the reaction pathways leading to the production of both nuclides, these two seem to be most important in the energy range investigated. The yield ratios for the production of ^{46}Sc and ^{47}Sc from vanadium have been found to be about 0.2. The reaction which gives rise to ^{48}Sc is about 3—17 per cent as possible as that yielding ^{46}Sc, and is about 0.6—4 per cent as possible as that yielding ^{47}Sc in the energy region from 45 to 60 MeV. The reaction leading to the production of ^{48}Sc from vanadium corresponds to that of ^{47}Ca from titanium. The yields of these reactions can be considered to be low. It appeared unlikely, just as in the case of the titanium target, that any ^{44m}Sc was produced in the irradiation of vanadium.

For analytical purposes, the lower limits of detection under the present experimental conditions were estimated. One mg of titanium gives an activity under the 0.511 MeV photopeak of 4.4×10^4 cpm,[24] so the lower limit of the titanium determination can be set at 1 μg. For vanadium, the limits of detection of 2 mg from the 1.31 MeV peak of ^{48}V and 60μg from the 0.160 MeV peak of ^{47}Sc are obtained.

22) The relative yields reported by Pai *et al.*[14] for the $^{49}Ti(\gamma,p)$ ^{48}Sc, $^{50}Ti(\nu,pn)^{48}Sc$ and $^{50}Ti(\gamma,pn)^{48}Sc$ reactions are 1.5, 0.17 and 0.37, respectively, at 30 MeV.

23) The same order in yield values was obtained in the irradiation of titanium with 20 MeV bremsstrahlung.[2]

24) The corrected value at the end of one-hour irradiation with 60 MeV bremsstrahlung (average beam current, 45μA): The geometry is 10% with a 3″ dia. × 3″ NaI(Tl) crystal.