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PHOTONUCLEAR REACTIONS IN CARBON AT ENERGIES BETWEEN 300 MeV AND 1000 MeV

V. di Napoli, F. Dobici, O. Forina, F. Saletti
(Laboratorio di Chimica delle Radiazioni e Chimica Nucleare del C.N.E.N., Istituto di Chimica Generale ed Inorganica - Università di Roma)

and

H. G. de Carvalho
(Centro Brasileiro de Pesquisas Físicas, Rio de Janeiro)

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ABSTRACT:

In the energy range from 300 MeV up to 1000 MeV, cross-sections for the reactions $^{12}C(\gamma, n)^{11}C$ and $^{12}C(\gamma, x)^{7}Be$ have been measured using bremsstrahlung beams of the Frascati electronsynchrotron by means of the activation method. The (γ, n) cross-section measured, 1.18 ± 0.20 mbarn, is in agreement with the predicted from the photomeson model. The $^{12}C(\gamma, x)^{7}Be$ cross-section found was $8 \pm 4 \times 10^{-29}$ cm^2.

INTRODUCTION

Inelastic interactions between complex nuclei and nucleons with energies in excess of 300 MeV have been extensively studied and the details of these processes are by now well known [1]. However, knowledge of inelastic interactions between complex nuclei and high energy photons is incomplete. This field has not yet been explored conveniently and the results presently available give only partial information. The present paper is part of a systematic work initiated to study in detail the inelastic interactions of high energy photons with complex nuclei. We intend to establish the magnitude, energy and mass number dependence of the photo-nuclear absorption cross-section at energies above the meson production threshold, since this is basic for understanding the interaction of high-energy quanta with complex nuclei. In this paper we report the (γ, n) and $^{12}C(\gamma, x)^{7}Be$ cross-sections from carbon for photons of energies between 300 MeV and 1000 MeV.

In this field the following papers have been published: Barber et al. [2], and recently Masaike [3] reporting cross-sections for photoreactions of ^{27}Al, Cu and ^{12}C, measured by the activation method, in the energy range from 150 to 720 MeV and Nefkens et al. [4], concerning production of ^{7}Be from Be, C and O, up to 270 MeV. The majority of previous studies on high energy photo-nuclear reactions have mainly involved the observation of emitted pions and nucleons rather than observation of residual nuclei. The meson production yield has a mass number dependence. Recently McClelland [5] found the dependence from 1000 MeV bremsstrahlung beams to be proportional to $A^{3/4}$.

Nuclear emulsion, from the study of photostars production, has shown a sharp rise above the meson threshold for the star production cross-section [6]. Debs et al. [7], Reagan [8] and Gorbunov et al. [9], have used the activation method to study some high energy photon interactions with complex nuclei. Debs et al., [7] irradiated elements of medium weight with 320 MeV bremsstralung and measured the relative yield of the products of reactions from high energy photons. Their results show a yield of products similar to the spallation produced by high energy nucleons in complex nuclei. Recently di Napoli et al., [10] obtained analogous results from the irradiation of Mn with 1000 MeV Bremsstrahlung. Reagan [8] measured the yield of ^{17}N from some elements as function of electron energy using delayed neutrons.

The activation method used in the present work offers advantages, the activities of residual products allow measurements of cross-section for individual process. When there is a yield of a stable residual product, it is possible from the yield of the radioactive products to assess the amount of the stable product. The magnitude of the total cross-section may be obtained by the sum of the cross-sections and gives important information regarding the mechanism of the process of interaction of photons with the complex nuclei. The mechanism of high energy photon interaction with the complex nucleus is mainly via meson production, related, therefore, to the total meson production cross-section. Thus, in principle, provided that the cross-section measurements are accurate enough, the activation method is capable of showing the resonance state of the pion-nucleon system effect, which appears as bumps on the total pion production cross-section curve. These bumps appear at the same energies [3] also in the cross-section curve of some complex nucleus reactions. At much higher energy where the total photo meson production cross-section is unknown, the cross-section energy dependence would indicate the shape of the total meson cross-section production curve. Therefore, this constitutes an indirect method of studying the energy dependence of photomeson production. The activation method is very useful for measurement of reactions in which photons are absorbed in the nucleus and only charged pions are emitted. Dyal and Hummel [11] for instance, have measured the cross-section for reactions such as $^{11}B(\gamma, \pi^-)^{11}C$ and $^{11}B(\gamma, \pi^+)^{11}Be$ and found the ^{11}C production several times larger than the ^{11}Be.

In 1953, Jones and Terwilliger [12] measured the photo-neutron production excitation function from 13.5 MeV to 320 MeV. The method used does not allow identification of the reaction from which neutrons have been emitted, therefore it is improper for measurements of the (γ, n) reaction cross-sections but gives an upper limit for them. Jones and Terwilliger assumed a quite high neutron multiplicity to explain the large cross-section (per neutron) which they found. The photon interacts with the nucleus via meson production and the subsequent meson absorption by nuclear matter leads to high excitation energies. In the case of uranium, photons of energy higher than 140 MeV would be capable of causing an evaporation of 9 neutrons. Recently, Bishop et al.[13] measured photo-neutron yields from C, Al and S by bremsstrahlung up to 200 MeV. Their results are larger than those of Jones and Terwilliger and show also photoneutron cross-section curves with a flat part extending from 35 MeV to the meson-threshold. Above 140 MeV there is a gradual increase of the cross-section. The pionic process contribution in this energy range (up to 200 MeV) was evaluated by Bishop et al. [13], and found to be less than 2%.

Determination of the cross-section by the photon difference method necessitates accurate measurements of the cross-section per equivalent quantum σ_Q at several energies. The radiochemical method is entirely adequate for measurements of some (γ, n) cross-section reactions. Carbon was chosen because of its low mass number and because it is almost monoisotopic and yields, from (γ, n) and $^{12}C(\gamma, x)^{7}Be$ reactions, products convenient for the activity measurements regarding mean-lives and the nature of radioactivity.

EXPERIMENTAL METHOD

The bremsstrahlung beam was obtained from the collision of electrons accelerated to the desired energy, with a thin tantalum target (0,05 cm). The dose measurements was always carried out by means of a quantometer of the Wilson type [14]. The constant used was 4.79×10^{18} MeV/Coulomb, following Gomez et al. [15].

The beam was "sharply" collimated so that it hit our target with a small cross-sectional diameter (1 cm). The possible ion-recombination within the quantometer (in the case of short pulses), was tested by comparing quantometer readings with the radio-activity simultaneously induced in copper discs at varying pulse lengths. No detectable effect was found within 0.2%. Inter-calibration was also made (for "long" pulses only) between the quantometer readings and those of a pair spectrometer: the comparison turned out to be satisfactory, a given dose being reproducible to better than 1%. The same pair spectrometer was used for determining to what extent the gamma ray absorption by an interposed thickness of material would alter the quantometer readings. Quantometer and spectrometer data were compared using various thickness of the target. [16]

To select electron energy the method chosen was to cut out, suddenly, the radiofrequency. Acceleration time was accurately measured by displaying a pattern given by a scintillation counter in an oscilloscope, and simultaneously triggering a calibrated oscillator whose pulses appeared super-imposed on the same screen. The polaroid photograph of this image is the energy measuring device. The time resolution in such photography is about 100 μsec., which corresponds to 1/200 of the total acceleration time at 1000 MeV. The energy measurement has an error of about ± 5 MeV at 1000 MeV and ± 15 MeV at 300 MeV.

Besides the high-energy photon reaction two sources of spurious reaction have to be considered.

A general background of diffused neutrons, as well as a neutron beam possibly generated in the tantalum target of the electrosynchrotron and in the collimator, must be considered as possible sources of spurious (n, xn) events. This effect was investigated by means of uranium-loaded nuclear emulsions used in fission cross-section measurements [16] but did not show an appreciable contribution from such neutrons. Secondaries from interactions within the target were also shown to be negligible both by simple calculation and by comparing yields in targets of different thickness.

The targets irradiated with the Frascati electronsynchrotron were polyethylene foils 8.4×10^{21} carbon atoms/cm^2 exposed at normal incidence to the beam. The targets were irradiated at constant beam intensity for a few minutes at five different peak bremsstrahlung energies in the range 0.3 GeV to 1 GeV, chosen so as to give four equal $\Delta \ln E$ intervals.

TABLE I

Energy MeV	Cross-sections per equivalent quantum (cm^2)	
	Beryllium	Carbon
300	$(4.0 \pm 0.6)\ 10^{-28}$	$(4.12 \pm 0.20)\ 10^{-27}$
400	$(4.3 \pm 0.6)\ 10^{-28}$	$(4.67 \pm 0.20)\ 10^{-27}$
550	$(4.5 \pm 0.6)\ 10^{-28}$	$(4.87 \pm 0.20)\ 10^{-27}$
750	$(4.7 \pm 0.7)\ 10^{-28}$	$(5.05 \pm 0.20)\ 10^{-27}$
1000	$(4.96 \pm 0.17)\ 10^{-28}$	$(5.50 \pm 0.20)\ 10^{-27}$

RESULTS AND DISCUSSION

$^{12}C(\gamma, n)^{11}C$ results - The results of the cross-section measurements are summarized in Table I and in figure 1 where both the cross-sections per equivalent quantum σ_Q are plotted as a function of the natural logarithm (ℓn E) of the peak bremsstrahlung energy. The slopes ($d\sigma_Q/d\ell n$ E) of the curve which best fit the experimental points corresponds to σ_k, the cross-sections at each photon energy. Within our experimental errors (10%) the (γ, n) cross-section in the energy range appears to be constant; Masaike [3] also found a constant (γ, n) cross-section from 200 MeV to 700 MeV. The (γ, n) cross-section was computed in first approximation using the "square" form of the bremsstrahlung spectrum $\sigma_k = (d\sigma_Q/d\ell n$ E) and then corrected for the actual form of the spectrum. The σ_k found was $(1.18 \pm 0.20) \times 10^{-27}$ cm^2. To verify the accuracy of our measurements the cross-sections per equivalent quantum σ_Q was computed by means of the numerical integral:

$$\sigma = \frac{1}{Q}\int_0^E \sigma_k \, n(k, E)dk \qquad (1)$$

The "number of equivalent quanta" Q is the ratio of total energy in the beam ε measured with the Wilson quantometer to the peak bremsstrahlung energy E, ($Q = \varepsilon/E$). In this case Q is normalized to one equivalent quantum and n(k, E) is the number of photons in the energy interval (k,k+dk) of the bremsstrahlung beam. The computation of the integral is possible when σ_k (the (γ, n) cross-section at each energy k) is available. To compute such a numerical integral we selected the following data: results of Miller et al.[17], Fultz et al. [18] and Cook et al., [19] in the giant resonance region. The results of Fultz et al. were obtained from monochromatic gamma rays and cover the energy range from threshold to 37 MeV. The results of Cook et al. [19] obtained by means of a new method of analysis of bremsstrahlung data, are somewhat larger, but follow very closely the same pattern structure as the cross-sections measured by Miller [17] and Fultz [18] and cover the range up to 65 MeV. Figure 6 shows these (γ, n) cross-sections. In the giant resonance, the Fultz et al. results are represented by a solid line; while a dotted line represents the Miller et al. results. From 37 MeV up to 65 MeV (solid line) we used the results of Cook et al., normalized to those of Fultz at 37 MeV. The (γ,n) cross-sections in the energy range from 65 MeV to 200 MeV are not accurate. Fortunately, since the cross-sections are very small, the contribution to the numerical integral is not larger than 10% in that energy range. We observed that, in the energy range from 35 MeV up to 65 MeV, (γ, n) cross-sections are nearly proportional to the deuteron photodisintegration cross-section. We assume that this is true up to 100 MeV, after which it tends to level off, passing a minimum value at 140 MeV and increases gradually to match our experimental values at 300 MeV, as shown in figure 2 by a dotted line. Once those cross-sections were selected, the numerical integrals were computed for 320 MeV and 1000 MeV bremsstrahlung spectra. (See figure 2). Assuming that the values of cross-sections used by us in the range of 65 MeV to 200 MeV are imprecise up to ± 50%, our results are in agreement with the absolute values of the numerical integrals within 10%. Masaike's[3] results are in agreement with the results of Barber et al.[2] and disagree with our numerical values (see figure 2) as found for the integral, even if the cross-sections were assumed to be zero from 65 MeV up to 200 MeV. However, when multiplied by a 1.2 factor, his results are in excellent agreement with ours over the energy range that overlaps our present measurements. Probably one of the two sets of results is affected by a systematic error. Fuchs and Lindemberger[20] and Cumming et al.[21] pointed out as one cause of frequent systematic error in the $^{12}C(p, pn)^{11}C$ cross-sections measurements, the thinness of the plastic foils used. Thin plastic foil loses ^{11}C by diffusion process; for this reason some cross-sections for $^{12}C(p, pn)^{11}C$ reported in the literature are lower than the actual values. For our targets thickness (167 mg/cm^2) this corresponds to a correction of 2,5%.

At energies above 300 MeV it is believed that the (γ, n) predominant process results from the following two reactions:

$$\gamma + n \longrightarrow p + \pi^- \qquad (2)$$

$$\gamma + n \longrightarrow n + \pi^0 \qquad (3)$$

The total photo-pion cross-section per nucleon in complex nuclei has been calculated from the cross-sections of free nucleons, taking into account the nucleon motion in the nucleus[3, 22]. The probability $P_{n\pi}$ that only produced mesons and recoiled nucleons escape from target nucleus without producing scattering or meson absorption, can be calculated as a function of the phonon energy and mass number of the target nucleus from mean-free paths of nucleons and mesons in nuclear matter [22]. For carbon the (γ, n) cross-section is given by:

$$\sigma_k = 6\, P_{n\pi}(k) \left\{ \sigma_{\pi^0}(k) + \sigma_{\pi^-}(k) \right\} \qquad (4)$$

The average value of $\bar{\sigma}_k$ in the energy range considered, calculated from equation (4), is 1.34 mb. The average experimental value found is 1.18 ± 0.20 mb, which is 12% lower than that predicted from the photon-meson model, and therefore, within our experimental error, in good agreement. The calculated and the experimental values are shown in fig. 2 for comparison. It is worthwhile to point out that for Iodine, di Napoli et al. [23] found, for the (γ, n) cross-section, values which are in desagreement with the prediction of the photomeson model, thus the anomaly seems to be observable only for heavy nuclei.

Beryllium yield results - The ^{7}Be is produced mainly from the following photon interactions:

$^{12}C(\gamma, \alpha n)^{7}Be$ with a Q of 26,3 MeV

$^{12}C(\gamma, d, t)^{7}Be$ with a Q of 43.8 MeV

$^{12}C(\gamma, 2p, 3n)^{7}Be$ with a Q of 54.5 MeV

The target irradiation time for ^{7}Be production was 12 hours. The half-life of ^{7}Be is 53 days, the electron-synchrotron bremsstrahlung beam intensity is not high enough to induce high activity, consequently the target activity was followed for more than one month. The activity decay in this period corresponds to a half-life of 53 days. The ^{7}Be 0.47 MeV photo-peak area measure ment was started only when the ^{11}C 0.51 MeV annihilation peak had been attenuated to negligible values. Chemical identification of ^{7}Be was also achieved by extracting it from a solution of the polyethylene target in C Cl_4 by means of a 1N HCl solution. The beryllium was then precipitated, with a few mg of $BeCl_2$ added carrier, by NH_4OH and $(NH_4)_2CO_3$.

Artus [24] has measured the ^{7}Be cross-section photoproduction from ^{12}C, using bremsstrahlung beams, from threshold up to 57 MeV and found an integrated cross-section of 6.0 MeV m barns. Nefkens et al. [4] measured the ^{7}Be photoproduction from ^{12}C from threshold up to 275 MeV. Artus [24] et al [4] found a cross-section maximum, located near 35 MeV, for the $^{12}C(\gamma, n)^{7}Be$ reaction, (see figure 3) but with quite different values.

The results of the ^{7}Be yield from interaction of photons with carbon from 300 MeV to 1000 MeV are shown in figure 1 and Table 1. Figure 4 shows the σ_Q computed from equation (1) using Artus [24] results up to 50 MeV and making the assumption shown in fig. 4 regarding the cross-section values in the energy range from 60 MeV to 200 MeV. The experimental points of Nefkens et al. are plotted for comparison. Our value at 300 MeV is a little larger than that of the Nefkens et al. [4] at 275 MeV but agree well, within the experimental errors.

The yield ratio ($^{11}C/^{7}Be$), from high energy proton interaction with ^{12}C, in the considered energy range, is nearly constant and equal to 3 [21]. Therefore the high energy proton going through the carbon nucleus has a cross-section for the ejection of neutron and a proton 3 times larger than to produce a residual ^{7}Be nucleus through one of the possible reactions. In the energy range considered, the yield ratio ($^{11}C/^{7}Be$) due to photon interactions as found in the present work, is quite different nearly 14. The reason is that ^{11}C and ^{7}Be are produced by photons in a different situation. The recoil nucleons and pions are uniformly originated in the bulk of the nucleus. The mean free paths of those particles are therefore 1/2 of the values for particles crossing the whole nucleus. The transparencies are thus twice the ordinary values. The overall effect for mesons and recoil nucleons is a decrease by a factor of 4 in the probability of the ^{7}Be formation.[20] Therefore the yield of ($^{11}C/^{7}Be$) = 14 seems to be expected just from those simple considerations.

The $^{12}C(p, pn)^{11}C$ reaction has been largely used as a monitor reaction for measurement of high energy proton beam fluxes. It is convenient to discuss the possible use of $^{12}C(\gamma, n)^{11}C$ instead of the copper $^{63}Cu(\gamma, n)^{62}Cu$ reaction for monitoring of high energy photon beams, provided the σ_k or σ_Q cross-sections for $^{12}C \longrightarrow {}^{11}C$ have been accurately determined. An ideal monitor reaction should yield a product with a half-life longer than the longest irradiation time to be monitored. ^{11}C has a half-life of only 20.4 minutes; therefore its use is limited by this factor. It is useful only in short irradiations, to calibrate a secondary monitor such as an ion chamber which is then used for longer irradiation. However, since the half-life ^{11}C is twice the half-life of ^{62}Cu

(9.9 minutes) which has been widely used as a monitor, the ^{11}C has at least equal possibility of use. There are other possible monitors for high energy photon beams. The ^{18}F of (γ, n) from fluorine is one with good perspective value since its half-life is 111 minutes. The other possible radionuclides useful as monitors for high intensity beams are ^{24}Na and ^{7}Be, with half-lives of 15 hours and 53,6 days obtained from irradiations of targets such as for example ^{27}Al (γ, 2pn)^{24}Na and ^{9}Be(γ, 2n)^{7}Be.

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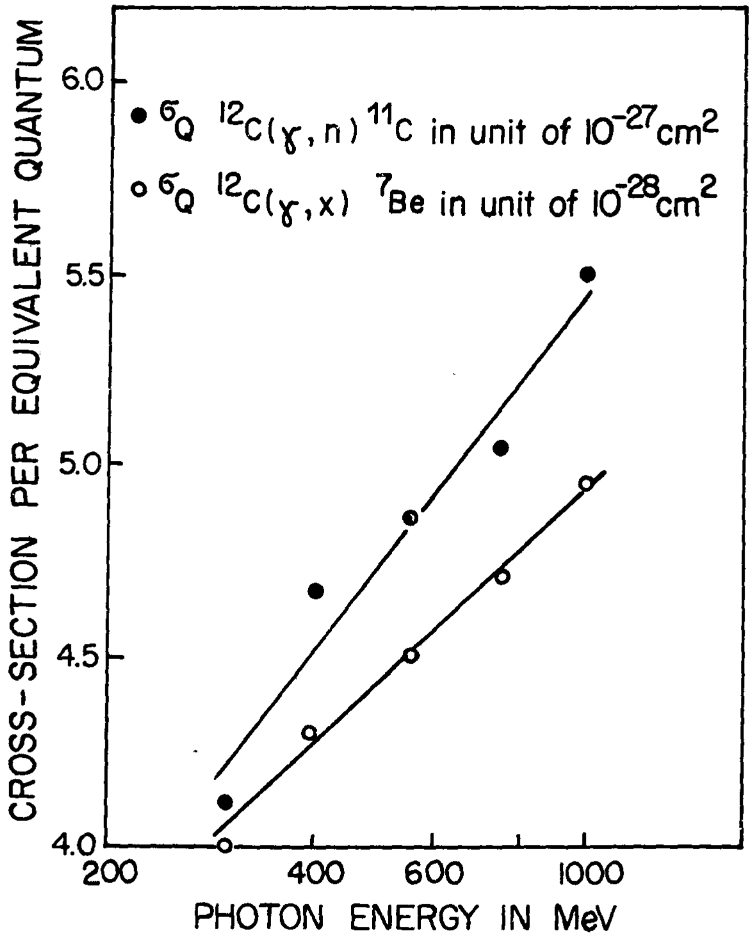
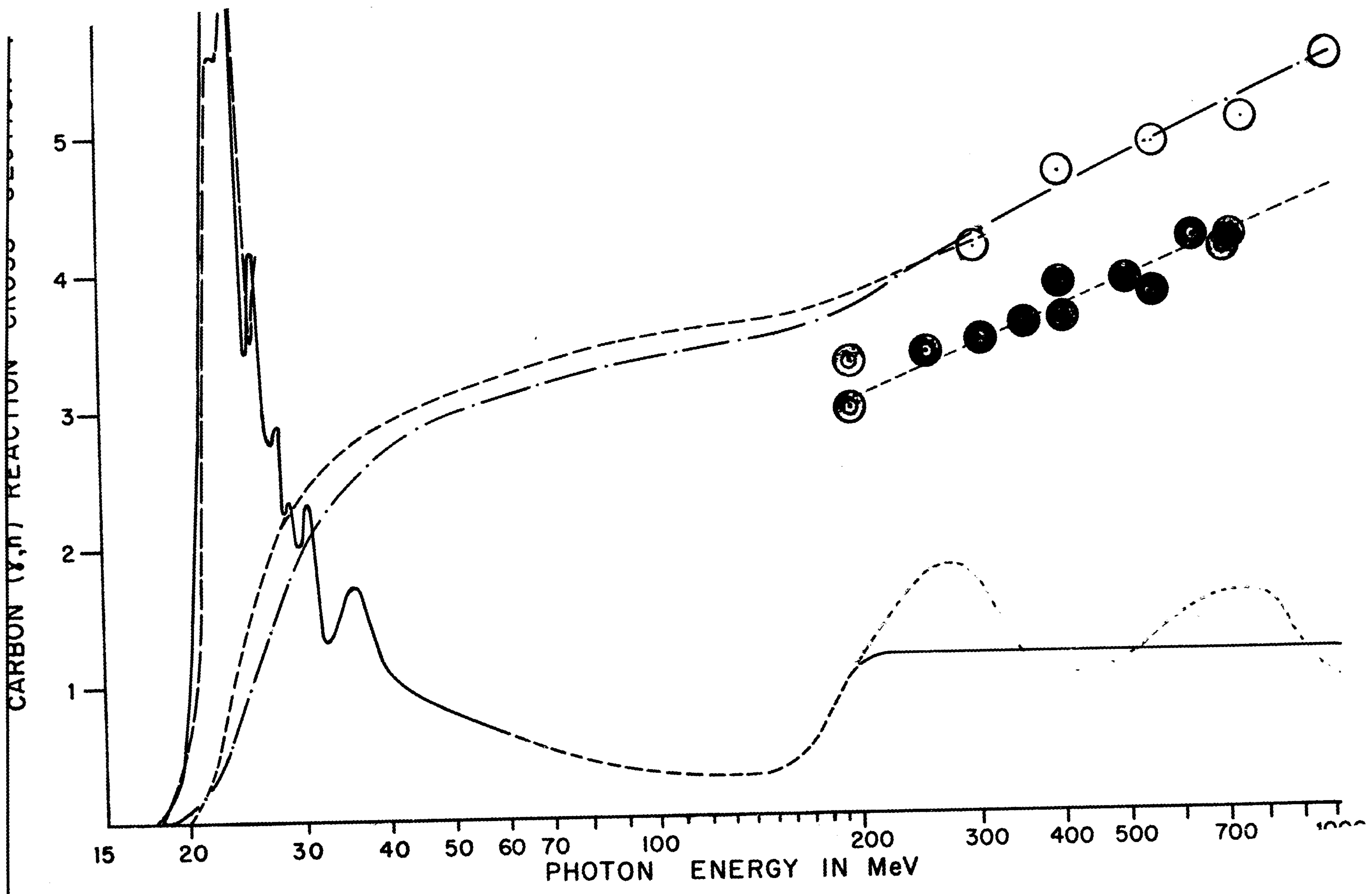
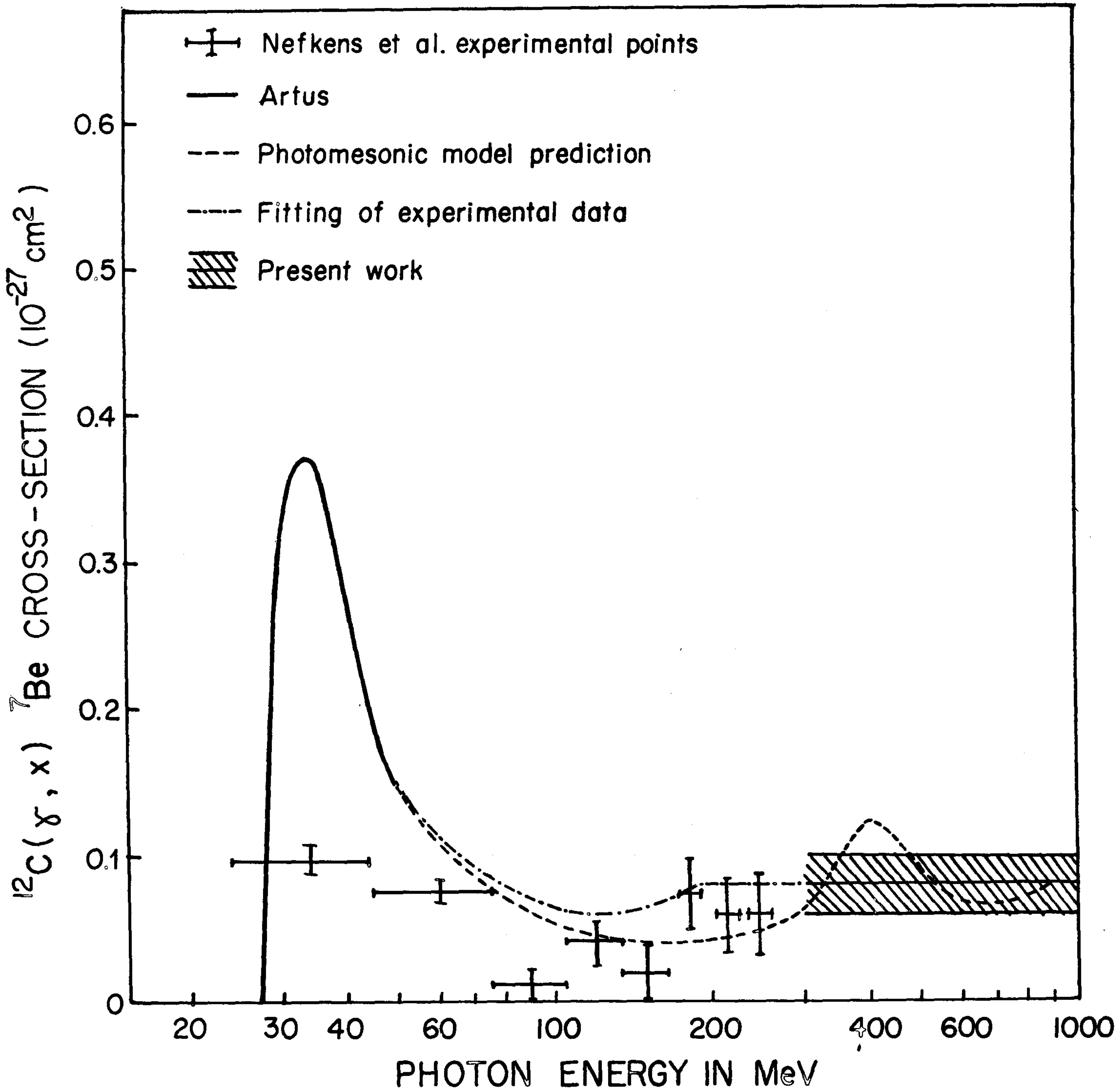


CROSS-SECTION PER EQUIVALENT QUANTUM
6.0
5.5
5.0
4.5
4.0
● σQ 12C(γ, n) 11C in unit of 10-27cm2
○ σQ 12C(γ, x) 7Be in unit of 10-28cm2
200
400
600
1000
PHOTON ENERGY IN MeV

FIG. 1


CARBON (γ,n) REACTION CROSS SECTION
5
4
3
2
1
15
20
30
40
50
60
70
100
200
300
400
500
700
PHOTON ENERGY IN MeV



Nefkens et al. experimental points
Artus
Photomesonic model prediction
Fitting of experimental data
Present work
$^{12}C(\gamma, x)$ ^{7}Be CROSS-SECTION (10^{-27} cm^2)
0.6
0.5
0.4
0.3
0.2
0.1
0
20
30
40
60
100
200
400
600
1000
PHOTON ENERGY IN MeV

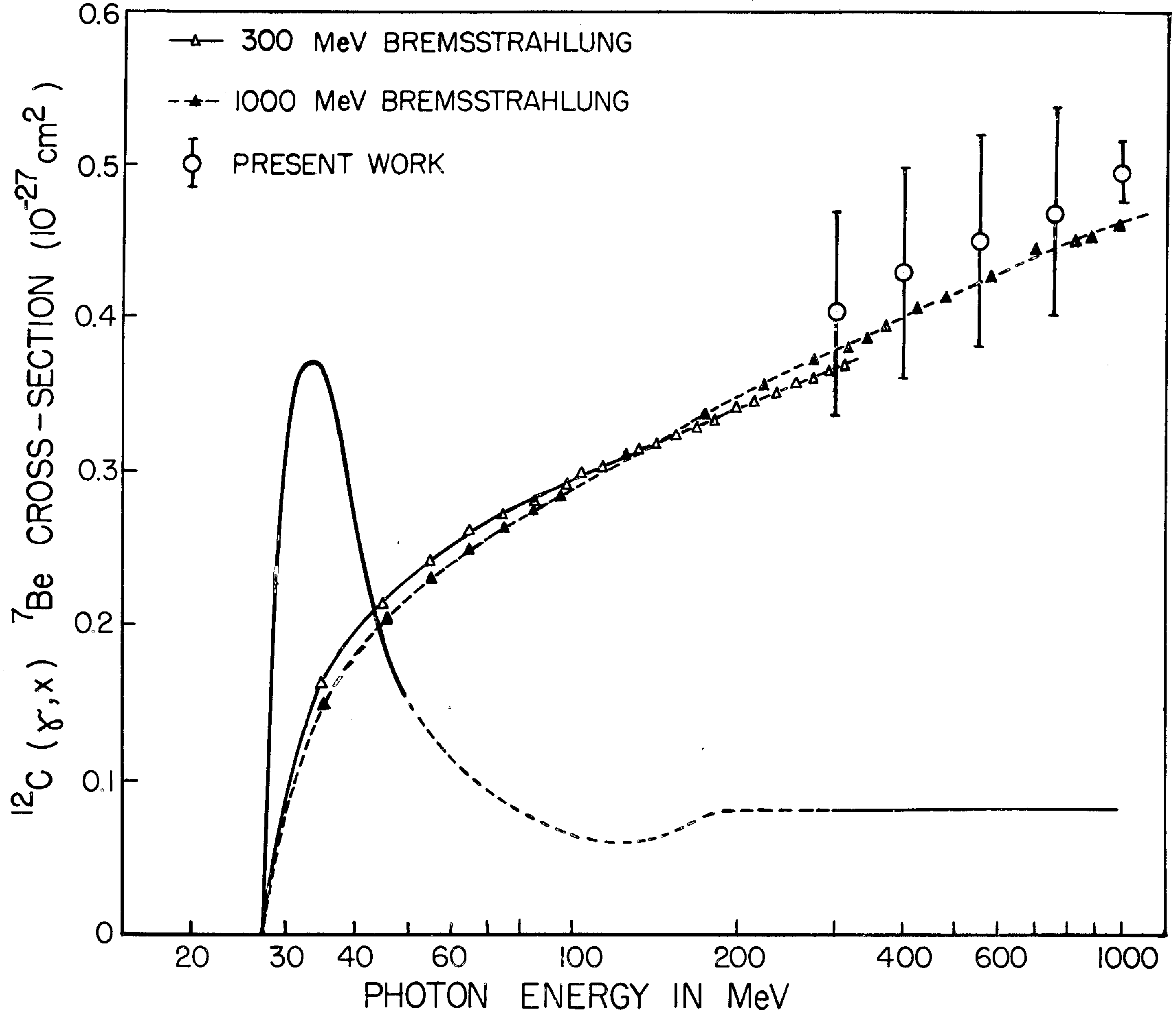

300 MeV BREMSSTRAHLUNG
1000 MeV BREMSSTRAHLUNG
PRESENT WORK
$^{12}C\ (\gamma, x)\ ^{7}Be$ CROSS-SECTION ($10^{-27}cm^2$)
0.6
0.5
0.4
0.3
0.2
0.1
0
20
30
40
60
100
200
400
600
1000
PHOTON ENERGY IN MeV

FIG. 4

FIGURE CAPTIONS

Fig. 1 - Experimental data for $^{12}C(\gamma, n)^{11}C$ and $^{12}C(\gamma, x)^{7}Be$ reactions cross-sections per equivalent quantum are plotted versus natural log of the peak energy of the bremsstrahlung.

Fig. 2 - Carbon (γ, n) cross-sections. Giant resonance data from Miller et al. [17] dotted line, and Fultz et al. [18], solid line. Cook et al. [19] from 37 MeV up to 57 MeV. From 57 MeV (dashed line) see text. From 300 MeV up to 1000 MeV obtained from our measurements (solid line). The dotted line represents results calculated from the photo-meson model. The numerical integrations, taking in account the actual shape of the bremsstrahlung spectra at 320 MeV and 1000 MeV, are shown to be in substantial agreement with the 1.18 mb cross-section found for the (γ, n) reaction in this energy range. Masaike's [3] results are shown for comparison.

Fig. 3 - $C^{12}(\gamma, x)^{7}Be$ cross-sections. Giant resonance data from Artus [24]. The quasideuteron region cross-sections were selected from Nefkens [4] et al. data adjusted to match our results at energies above the meson threshold. Our results from 300 MeV up to 1000 MeV are reported by the dashed area. The dashed line represents the cross-section calculated from the photo-mesonic model normalized to our average result.

Fig. 4 - Shows the computation of the σ_Q, cross-sections for equivalent quantum, at 320 MeV and 1000 MeV, from the σ_k, the cross-section at each photon energy, by means of the numerical integration using the actual shape of the bremsstrahlung spectra (see text). Our experimental points are shown for comparison.

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