EXCITATION FUNCTIONS OF SOME

MONITOR REACTIONS

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Samuel Meghir

EXCITATION FUNCTIONS OF SOME MONITOR REACTIONS

ABSTRACT

The excitation functions of the reactions Al²⁷(p,3pn)Na²⁴ and Cu⁶⁵(p,pn)Cu⁶⁴ have been determined relative to that of $C^{12}(p,pn)C^{11}$ from 20 - 90 Mev.

The activities of C^{11} , Na²⁴ and Cu⁶⁴ were measured by various techniques. The deviations in the cross sections ranged from \pm 6% to \pm 11%, depending on the accuracy of the technique employed.

Final results were compared with those previously reported in the literature. In cases where disagreement occurred, it has been possible to explain the discrepancy in terms of secondary reactions arising from the irradiations in some of the previous work.

INTRODUCTION

I GENERAL

Nuclear reactions have received ever increasing attention as a tool of research in the field of nuclear science. It is generally believed that a quantitative understanding of the mechanisms by which they proceed may lend the key to the solution of the complicated problem of nuclear structure.

A nuclear reaction is defined as a process in which a nucleon (projectile) impinges on a nucleus (target) imparting to it part or all of its kinetic energy. A number of single particles, or clusters of them, are subsequently knocked off the excited nucleus, thus leading to the formation of new residual species.

The probability of a particular type of nuclear reaction occurring at a certain bombarding energy is known as the 'cross section' of that reaction. This probability can be experimentally determined at any bombarding energy by measuring the number of product nuclides formed during the course of the particular reaction under study. The equation relating these two quantities is the following:

$$\frac{dN_p}{dt} = In_t \sigma_p \qquad (1)$$

where $\frac{dN_p}{dt}$ is the rate of production of the product nuclides, I is the intensity of the beam of bombarding particles or 'flux' expressed as the number of particles per cm^2 per sec. This treatment assumes that neither the energy nor the intensity of the beam is appreciably degraded. In this work the targets used were sufficiently thin to meet the requirements for the above conditions,

- nt is the number of target nuclei presented to the beam,
- op is the probability for the formation of the
 particular nuclide p under the conditions of the
 reaction, i.e. its cross section.

When the product nuclide is radioactive, then its decay during the time of bombardment must be taken into consideration. Thus the actual equation is:

$$\frac{dN_p}{dt} = In_t \sigma_p - N_p \lambda_p \qquad (2)$$

where $N_p \lambda_p$ is the rate of decay of the product nuclide, λ_p being its decay constant.

This equation can be easily solved for the number of nuclei found at the end of bombardment, N_p^o . The integrated form of equation (2) is:

$$N_{p}^{\circ}\lambda_{p} = In_{t}o_{p} (1 - e^{-\lambda_{p}t}) \dots (3)$$

where t is the length of bombardment.

If one takes into consideration that $N_p^{o\lambda}$ is the disintegration rate of the product nuclei at the end of bombardment, equation (3), if solved for O_p , becomes:

$$\sigma_{p} = \frac{(D.R.)_{p}}{In_{t}(1 - e^{-\lambda_{p}t})} \qquad (4)$$

The dimensions of the cross section, as it can be easily seen from equation (4), are those of an area. Physically, it represents the cross sectional area that the

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target nucleus presents to a beam of incoming particles.

The unit of cross section is the 'barn' - 1 barn = 10^{-24} cm².

II MECHANISMS

The mechanisms by which nuclear reactions take place depend largely on the energy and type of the bombarding particles. Various types of mechanisms have been suggested.

1. Compound nucleus formation

This mechanism was suggested by Bohr⁽¹⁾ and, briefly. it states that:

- (a) Any particle which hits the target nucleus is captured. This merely means that an incident particle will interact with the first one or two nucleons of the target transferring much of its energy to them, and thus to the nucleus, before penetrating it appreciably. Then it may no longer possess sufficient kinetic energy to escape, and thus a compound nucleus is formed.
- (b) The compound nucleus is excited to an energy state equal to the kinetic energy of the projectile plus the binding energy of the new particle. Complete equipartition of energy occurs among the nuclear constituents. It is also assumed that the lifetime of the compound nucleus is long ($\sim 10^{-12}$ to 10^{-14} sec) compared with the time required for a projectile of energy E to traverse the nucleus (10^{-21} sec). The

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long lifetime of the compound nucleus allows this redistribution of the energy acquired among the nucleons, in a statistical manner, until enough energy is accumulated on one of the nucleons to enable it to overcome the attraction of the nuclear forces and escape from the nucleus. An alternative mode of de-excitation of the nucleus is by emission of gamma rays.

(c) The disintegration of the compound nucleus is independent of its mode of formation. Therefore there will be a definite probability that it may decay into several possible residual nuclei. The disintegration by emission of nucleons is known as the 'evaporation step' of the mechanism and was theoretically treated by Weisskopf⁽²⁾, Le Couteur⁽³⁾ Naturally, the 'evaporation' can take and others. place only when the excitation energy of the compound nucleus is above the threshold for particle emission, and it will continue until all of its energy is dissipated. If the excitation energy is lower than the threshold for particle emission, then the compound nucleus will de-excite entirely by gamma-ray emission. However, the latter is a rather unlikely process when charged particles are used as projectiles, and it usually occurs when the target nuclei have low atomic number.

The experimental characteristics of such a mechanism

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applied in a nuclear reaction should be the following:

- (a) The angular distribution of the emitted particles should be spherically symmetrical, and their energy spectra should show a Maxwellian distribution.
- (b) The excitation function of any particular type of reaction should rise sharply above the threshold, pass through a maximum and then fall off rapidly as a new type of reaction becomes more probable.

The compound nucleus mechanism seemed adequate in explaining experimental data for the low energy particles used initially (up to about 30 - 40 Mev). However, it broke down when projectiles of higher energies became available for the study of nuclear reactions.

2. Serber mechanism⁽⁴⁾

This mechanism is applicable at somewhat higher energies. The basic idea is that the interaction between a projectile and a target nucleus depends on the way this interaction takes place and the amount of time it takes the incoming particle to traverse the nuclear field.

It has been shown that the mean free path of a particle traversing nuclear matter increases with energy. Serber estimated that the mean free path for a 100-Mev nucleon is 4×10^{-13} cm. He assumed that the momentum transfer does not increase proportionally with increasing projectile energy. He estimated it to be about 25 Mev per collision^{*} approximately

^{*}The word 'collision' does not necessarily imply physical contact between an incoming particle and a target nucleon. In reality, 'collision' is an interaction with transfer of energy and/or charge. The term 'collision' is used here in order to give a physical picture of the mechanism discussed.

for 100-Mev projectiles. Hence, when an incident particle traverses the nucleus, it may undergo only one collision before escaping it, having lost about 25 Mev of its energy. It may also undergo a small number of collisions, thus causing the emission of a number of nucleons before escaping, or even share all of its energy with the target nucleons. This step, which results in a prompt emission of nucleons, is usually referred to as the 'cascade' step of the mechanism. Since the struck nucleons have much lower energies than the incident particle and shorter mean free paths, they can escape from the nucleus only if the collision occurred near its edge; otherwise they will stay within the nucleus, sharing their energy with other nucleons. The subsequent events can then be described in terms of evaporation theory (2,3) whereby the residual energy imparted to the struck nucleus is dissipated by successive emission of particles, each carrying a few Mev of energy. The following conclusions may be drawn by applying the Serber mechanism to nuclear reactions:

(a) Because of the wide distribution of excitation energies of the struck nuclei, a variety of residual nuclei is expected after the evaporation processes are completed.
(b) The cross sections of nuclear reactions should vary slowly at high energies, since the energy transfer does not vary greatly.

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[&]quot;Here too the term 'nucleus' should not be taken literally to mean a solid mass with a well-defined boundary. It should rather be thought of as consisting of a hard core containing the nucleons in closed shells and a cloud of n nucleons in open shells all held together by some exchange forces, but with no sharply defined boundary(11).

(c) The total cross section (i.e. the sum of cross sections for all possible interactions for absorption or scattering) should be closer to πR^2 (R = nuclear radius) for heavier nuclei than for lighter nuclei at high energies.

(d) The density of emitted particles should be higher in the forward direction in the fast cascade step.

The applicability of Serber's mechanism was tested by a large number of experiments (e.g. 5,6,7,8,9,10) and it was found that it fits best to experimental data above 100 However, even at lower energies, this mechanism cannot Mev. be excluded. Actually, as it was shown by Weisskopf⁽¹¹⁾, both mechanisms exist. The prevalence of either one depends on the energy of the incoming particles. At lower energies the compound nucleus mechanism is predominant. However, as the energy of the incoming particles increases, there is a gradual transition which culminates in a complete Serber mechanism for the energy region of 100 Mev and above. Furthermore, when the study of nuclear reactions was extended to more complex nuclei and higher bombarding energies, it was found that even the Serber mechanism could not explain the experimental results adequately. Increasing evidence pointed to the fact that, in more complex nuclei, light nuclides (deuterons, alpha particles and even heavier nuclei) as well as nucleons were emitted (e.g. 10,12,13,14,15,16).

The initial step of the Serber mechanism (fast cascade step) was first treated quantitatively by Goldberger⁽¹⁷⁾.

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He calculated nuclear reaction cross sections by following the passage of particles through nuclear matter, step by step, until, for a large number of cases, they either escaped from the nucleus or lost sufficient energy to be captured. Whenever a choice between equally probable events had to be made, the choice was made at random. The name 'Monte Carlo' that was given to this type of calculation originated from this randomness of choice.

The 'Monte Carlo' technique was further refined and, with the use of computers, it became possible to obtain results which, although still not satisfactory, were more accurate than those of Goldberger, since a larger number of events were studied (18,19). The evaporation step has also been treated in this manner (20,21,22), the calculations being based on the already well-known evaporation theory (2,3).

Further description of the 'Monte Carlo' technique is beyond the scope of the present work. Those interested in further details should consult reference 19.

III TECHNIQUES OF INVESTIGATION

Essentially there are four techniques by which nuclear reactions may be experimentally investigated.

- (1) Study of the type, number, energy, and angular distribution of the emitted particles.
- (2) Determination of the yields of the residual product nuclei by mass spectrographic means.
- (3) Direct study of individual nuclear reactions by means of nuclear emulsions.

The above three methods are the so-called physical methods, as no chemical separation of the products is required.

(4) The fourth method is the so-called radiochemical technique. It consists in measuring the activity of the residual radioactive nuclides formed by suitable means after chemical separations.

Since the difference of the atomic number of the nuclides is used in order to achieve a chemical separation, isotopes of the same nuclide cannot be separated. However, yields of individual isotopes can be measured in most cases if use of their different decay characteristics is made. Independent yields of radioactive nuclides, which are also produced by decay of some other parent isotope formed during a bombardment, can also be measured by utilizing suitable parent-daughter decay relationship, e.g. 'milking' a daughter from the parent.

Unless care is taken, chemical separations can be subject to large errors due to the minute amounts of product nuclei formed (10^{-12} gm.) . Addition of inactive carriers, however, makes possible the use of semi-micro and microchemical techniques of separation, and also the estimation of the amount of recovered activity in cases where separations are not 100% efficient. The radiochemical technique was used throughout this work.

The results obtained by methods (2) and (4) can be treated in two main ways in order to yield the desired

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information.

- (a) Study of the yields of nuclei of different mass number, produced at a defined bombarding energy.
- (b) Study of the variation of the yield of a particular isotope produced by a nuclear reaction at different bombarding energies.

The graphical representation of the yield versus energy of bombardment is called the 'excitation function' of the reaction. Method (b) is the most common in use.

IV SOURCES OF PROJECTILES

In the early studies of nuclear reactions, the only sources of projectiles available were those provided by nature. However, the limitations as to the type of particles they furnished, as well as the energy range of the particles, soon led to a search for some new and more versatile types of projectile sources capable of yielding particles of higher One of the most successful of them was the energies. 'cyclotron', a circular type of particle accelerator proposed and developed by E.O. Lawrence and his coworkers (23,24, 25,26) Refined models of the cyclotron are in use today, capable of accelerating many types of particles to energies as high as 30 Bev. The most common type in use today is the 'synchrocyclotron'. In this model the frequency of oscillations of the electric field is varied in order to take care of the apparent increase in mass of the accelerated particles (relativistic effect) which occurs when the latter

reach high velocities. This modification was proposed independently by McMillan⁽²⁷⁾ and Veksler⁽²⁸⁾ for the acceleration of electrons (McMillan's 'Synchrotron') and was successfully applied by Lawrence's coworkers to the cyclotron for the acceleration of protons and other heavier particles^(29,30,31).

V BEAM INTENSITY MEASUREMENT

From equation 4, P. 2, it is clearly seen that, in order to measure the cross section of a nuclear reaction, the beam intensity must be known, since all other quantities can readily be measured.

The methods by which the beam intensity can be measured generally fall into two categories:

(1) Absolute measurements.

In this category belong measurements done by various devices which take advantage of the charge carried by the bombarding particles. The charge can be determined either directly or by the ionization it causes. However, these devices require that an external beam be used. This means that they are good only when one energy measurement is done. If the reaction is to be studied at various energies by means of an external beam, the degradation is achieved by suitable absorbers. However, this method of varying the beam energy introduces problems of contamination by secondary particles and energy spread. In addition, the use of absorbers attenuates further a beam which may already be weak. Therefore, in many nuclear reaction studies, the internal beam of the accelerator (in the case of the present work the synchrocyclotron) is used. The energy variation can then be achieved by inserting the target inside the cyclotron at various distances from the centre. In this case, absolute measurement of the beam intensity is virtually impossible and the relative method must be used.

(2) Relative measurements.

This is done by using a monitor reaction which is a nuclear reaction whose cross section is accurately known. If the target and the monitor are combined in such a way that they present the same area to the beam, it may be safely assumed that they both receive the same flux (i.e. number of particles per cm^2 per sec) provided the combined target is thin enough so that there is neither appreciable attenuation of the beam intensity nor any energy degradation. By this means the beam intensity can be deduced from the monitor reaction, and the value may then be inserted in the equation for the target under study.

Any well-known nuclear reaction may serve as monitor, but the most common are the following:

> 1. $B^{11}(p,n)C^{11}$ 2. $C^{12}(p,pn)C^{11}$

3. $Cu^{63}(p,n)Zn^{63}$ 4. $Cu^{65}(p,pn)Cu^{64}$ 5. $Al^{27}(p,3pn)Na^{24}$ 6. $Al^{27}(p,3p3n)Na^{22}$

The range of usefulness of these nuclear reactions is determined by their threshold and shape of the excitation function.

Of the above nuclear reactions, Nos. 2, 4 and 5 have had the widest application.

VI LITERATURE REVIEW

1. $C^{12}(p,pn)C^{11}$

Chupp and McMillan⁽⁸⁾ investigated the crosssection variation for the production of C¹¹ by bombarding polystyrene foils with 140-Mev protons obtained by the stripping of deuterons. Their aim was to test the Serber mechanism. The beam they used was collimated and the degradation was achieved by means of carbon absorbers. The beam was monitored electronically. They reported that the cross section did not vary between 60 to 140 Mev, thus confirming Serber's mechanism.

McMillan⁽³²⁾ determined the value of the cross section at 62 MeV and found it to be 73 mb. Later McMillan and Miller⁽³³⁾ recalculated this value and brought it down to 69 mb.

Panofsky and Phillips (34) measured the excitation function of this reaction from the threshold up to 32 Mev by means of a linear accelerator. They were mainly interested in finding the threshold of the reaction and investigating the possibility that the reaction might be a (p,d) instead of a (p,pn). They reported a threshold of $18.5 \stackrel{+}{-} 0.3$ Mev, and their calculations, based on the mass number of B¹¹ and the energy of the β^+ emitted by C¹¹, proved that the reaction is a (p,d) one, at least near the threshold. They also reported a value of the cross section at 32 Mev to be 75 $\stackrel{+}{-}$ 2 mb. Their method of degrading the beam was essentially the same as that used by Chupp and McMillan⁽⁸⁾.

Hintz and Ramsey⁽³⁵⁾ quoted the results of Chupp and McMillan as incorrect due to their method of degradation of the beam. They pointed out that the use of the C absorbers produces a large number of secondary particles, namely neutrons, which have a relatively high cross section in producing C^{11} via a $C^{12}(n,2n)C^{11}$ reaction. They then redetermined the excitation function of the $C^{12}(p,pn)C^{11}$ reaction along with those of $Al^{27}(p,3pn)Na^{24}$ and $Al^{27}(p,3p3n)Na^{22}$.

They also used an external beam but with brass absorbers which, they believed, gave a much lower neutron background. Their brass absorbers were interposed between the carbon target foils, thus producing a full excitation function in one bombardment.

This technique, called 'stacked foil' technique, is fully described by Aamodt et al. (36) and was in wide use at the time. Hintz and Ramsey claim an error of $\frac{+}{-}3\%$ on the basis of the smooth fit they obtained when they compared their results to those of Aamodt et al. The latter (36)were the first to report a full excitation function curve for this reaction from the threshold up to 340 Mev. Actually the curve consisted of three parts: one from the threshold up to 32 Mev, which was obtained by using a linear accelerator, one from 93 up to 340 Mev, which was obtained by means of the Berkeley synchrocyclotron, and a third part between 32 and 93 Mev, which was that of Hintz and Ramsey after normalization at two energies, one at 110 and one at 32 Mev.

Aamodt et al. also used the 'stacked foil' technique and measured the beam intensity by a Faraday cup. They took into consideration the neutron background which, they claimed, was 2%. Their value at 32 Mev was $89 \stackrel{+}{=} 4$ mb, agreeing with that of Panofsky and Phillips⁽³⁴⁾, while McMillan and Miller's⁽³³⁾ value at 62 Mev was found to be lower.

A strange feature in Aamodt's results was a dip in the curve occurring at 340 Mev. Warshaw, Swanson, and Rosenfeld⁽³⁷⁾ checked this strange feature by determining the cross section at two energies, namely 283 Mev and 387 Mev. The cross-section values they reported were 48.9 ± 2.5 mb and 43.6 ± 2.2 mb respectively.

Later Rosenfeld⁽³⁸⁾ repeated the work at 400 Mev and found a value of 34 mb. This agreed with the results of Crandall et al.⁽³⁹⁾ which had recently appeared. He

therefore withdrew the previous values. Crandall et al. (39)measured the excitation function of $C^{12}(p,pn)C^{11}$ from 105 to 350 Mev in order to confirm or disprove the existence of the dip reported by Aamodt et al. They showed that no dip In view of the possible sources of occurred at 340 Mev. error that could have entered into Aamodt's experiments, they exercised great care in their own experiments. They too used an external beam and carbon absorbers to achieve the beam degradation, but they carried out two series of experiments, one with the absorbers directly in front of the target and one with the absorbers in front of the beam collimator. The second series was free of secondary effects. Their beam current was carefully checked by both an ionization chamber and a Faraday cup placed before and after the stack of target and absorber foils respectively. They also exercised great care in their activity measurement techniques. In fact. Seliger^(40,41) had recently found that the backscattering of positrons and negatrons differed, and thus the contributions to the different measurements by this effect had to be carefully reassessed. This source of error had not been taken into account in all previous results. Crandall et al. used 47 measurement techniques which did not involve backscattering, and they determined experimentally the selfabsorption correction for positrons emitted by C¹¹ in their particular case. On the basis of these factors, they found that, although the shape of the excitation function remained the same, Aamodt's results were high by about 13%. They

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explained the dip at 340 Mev by the fact that the secondary effects in this region are more pronounced, since the contribution comes from both secondary neutrons and protons, than in the lower energy region where the contribution comes mainly from neutrons, thus giving a net artificial rise to the cross section values obtained by Aamodt. They actually reproduced the dip when they repeated the work of Aamodt in the same way. At 340 Mev they found a cross section of 36 mb, in good agreement with that of Rosenfeld⁽³⁸⁾ at 400 Mev (34 mb) and a later value by Rosenfeld, Swanson, and Warshaw⁽⁴²⁾ who reported a cross section of 31.1 mb at 461 Mev.

The views of Crandall et al. (39) were fully supported by Rosenfeld et al. (42) who gave a thorough discussion of all possible sources of error responsible for previous disagreements in the cross-section values for the $C^{12}(p,pn)C^{11}$ and $Al^{27}(p,3pn)Na^{24}$ reactions. These errors include the following:

(1) Errors due to activity produced by secondary particles, especially neutrons, originating from the beam degradation. The effect is more pronounced in the case of the 'stacked foil' technique, since there is direct contact between target and absorber foils and therefore there is a considerable flux of neutrons which, in some cases, have quite high cross sections for reactions of the (n,2n) type. For the Al²⁷(p,3pn)Na²⁴ reaction, Rosenfeld et al. pointed out that a more probable reaction, yielding Na²⁴ at low energies, is the Al²⁷(n, a)Na²⁴, which also has a lower threshold than Al²⁷(p, 3pn)Na²⁴. Therefore care should be exercised when the 'stacked foil' technique is used in this case too.

(2) The absolute beta measurement technique can also introduce significant errors, especially where positron emitters are involved, unless proper precautions are taken. Besides the difference in backscattering coefficients, there may be a similar difference in self-absorption coefficients for negatron and positron emitters of the same maximum energy. Thus, they pointed out, there may still exist discrepancies of the order of 10% in reported values, originating from the methods of beam intensity measurements, preparation of calibration curves for activity measurement, and even the activity measurements themselves.

Whitehead and Foster⁽⁴³⁾ reported that the 13% correction quoted by Crandall et al. for the high energy part of the Aamodt results should be applied also in the lower energy region down to 32 Mev. Further evidence for the correctness of Crandall's results came in 1960 in a paper by Parikh⁽⁴⁴⁾ who determined the excitation function of the $C^{12}(p,pn)C^{11}$ reaction from 68 to 383 Mev, exercising great care in the beam collimation and decontamination from neutron background. While his targets were thicker than had been used by past investigators and he employed gamma counting

techniques instead of beta, his results agree well within experimental error with those of Crandall et al.

No further work was reported for the low energy region, especially below 100 Mev which is the energy range of the McGill Synchrocyclotron. The excitation function of the $C^{12}(p,pn)C^{11}$ reaction was, however, extended towards the higher energy region, up to about 6 Bev. (45,46,47,48,49,50)

Table I contains values available from the literature. Fig. 1 is a composite curve constructed on the basis of the data presented in Table I. The lower energy portion of the curve is the excitation function reported by Aamodt et al.⁽³⁶⁾ corrected according to Crandall et al.⁽³⁹⁾.

Table I

COLLECTED	CROSS	SECTIO	N VALUES	FOR	THE
(2 ¹² (p.)	$(n)c^{11}$	REACTION		

Energy in Mev	Beam Monitoring Techniques	Activity Measurement Techniques	Reported value in mb	Ref	Remarks
32	A	B(E)	75 ± 2	34	
32	A	B(E)	77.5 ± 4.0	36	a
50			86.9		Ъ
60			80.8		Ъ
62	A	B(E)	69 ± 7	33	
70			76.5		Ъ
80			70.4		b
90			66.0		Ъ
93	A	B(E)	61.3 ± 3.6	36	8.
110			57.2		b
125			51.7		Ъ
135			49.0		b

Energy in Mev	Beam Monitoring Techniques	Activity Measurement Techniques	Reported value in mb	Ref	Remarks
144	A	B(E)	49.1		 8
150		2(2)	44.6		Ъ
150	R(660)	в	46.2 ± 1.9	47	c
170	A (000)	Б(I)	39.7 ± 0.9	39	-
175		2(27	38.7		Ъ
194	A	B(E)	45.2 ± 1.5	36	8.
200		- (-)	36.3		ъ
204	A	B(I)	37 ± 2	39	
225			36.0		Ъ
238	A	B(E)	35.8 ± 2.4	46	đ
240	A	B(I)	37.2 ± 1.8	39	
245	A	B(E)	43.3 ± 1.2	36	a
250		- (-)	36.0		Ъ
260	R(660)	В	38.2 ± 0.6	47	с
263	A	B(E)	43.9 ± 2.6	36	8.
270	A	B(I)	35.9 ± 1.0	39	
275			36.0		Ъ
288	A	G	33.7 [±] 1.2	44	
290	R(660)	В	37.0 ± 1.2	47	c
293	A	B(E)	41.5 ± 1.0	3 6	8.
2 95	A	B(I)	35.5 ± 1.0	39	
2 95	A	B(I)	37.9 ± 0.4	39	
300			36.0		Ъ
310	A	B(E)	31.9 ± 2.4	46	đ
313	A	G	34.8 ± 1.1	44	
313	A	B(E)	41.4 7 2.1	3 6	8.
320	A	B(I)	35.5 - 0.7	39	
325	A	B(I)	35.9 ± 0.8	39	
339	A	G	34.9 ± 1.0	44	
340	A	B(E)	35.8 ± 0.5	36	8.
340	A	B-G coinc.	36.0 ± 3.0	39	
350	R(660)	В	36.0 <u>±</u> 1.2	47	C
350	A	B(I)	36.0 - 0.7	39	

Table I (Contd.)

Energy in Mev	Beam Monitoring Techniques	Activity Measurement Techniques	Reported value in mb	Ref
350	$R(Na^{24})$	B(E)	35.0	45
350	$R(Na^{24})$	B(E)	33.5	45
362	A	G	32.4 ± 1.0	44
365	A	B(E)	37.4 ± 3.1	46
383	A	G	31.6 ± 1.0	44
390	$R(Na^{24})$	B(E)	34.5	45
400		B(E)	34	38
420	$R(Na^{24})$	B(E)	33.5 ± 1.7	45
420	A	B(E)	32.3 ± 2.9	46
440	$R(Na^{24})$	B(E)	31.7	45
450	R(660)	В	32.0 ± 0.6	47
461	A	B(I)	31.1 ± 1.0	42
465	A	B(E)	29.4 ± 1.2	46
		- ()	+ - /	1. 1

Remarks

e

e

Table I (Contd.)

365	A	B(E)	37.4 ± 3.1	46	d
383	A .	G	31.6 ± 1.0	44	
390	$R(Na^{24})$	B(E)	34.5	45	e
400		B(E)	34	38	
420	$R(Na^{24})$	B(E)	33.5 ± 1.7	45	e
420	A	B(E)	32.3 ± 2.9	46	đ
440	$R(Na^{24})$	B(E)	31.7	45	e
450	R(660)	В	32.0 ± 0.6	47	c
461	A	B(I)	31.1 ± 1.0	42	
465	A	B(E)	29.4 ± 1.2	46	d
522	A	B(E)	33.2 ± 1.6	46	đ
560	R(660)	В	30.4 ± 0.6	47	c
600	$R(Na^{24})$	B(E)	27.5 ± 1.5	45	•
648	A	B(E)	25.5 ± 3.0	46	d
660	A	В	31.0 ± 1.7	47	С
832	A	B(E)	30.0 ± 1.7	46	đ
950	A	B(E)	23.4 ± 1.3	46	đ
1000	$R(Na^{24})$	B(E)	26.1 ± 2.1	45	e
1400	$R(Na^{24})$	B(E)	24.1 ± 3.0	45	e
1800	$R(Na^{24})$	B(E)	22.6 ± 3.8	45	e
2000	A	G	26.0 ± 0.9	48	
2200	$R(Na^{24})$	B(E)	23.2 ± 5.0	45	e
2950	$R(Na^{24})$	B(E)	22.0 ± 6.0	45	
3000	A	G	26 ± 1	48	
3000	A	G & B	29.8 ± 1.6	49	
4100			30.5 ± 4.1		f
4500	A .	Gł&B	27.7 ± 1.7	49	
5700	$R(Na^{24})$	G	29 ± 3	50	
6000	A	G & B	29.8 ± 1.6	49	

Table I - Explanation of Symbols

Beam Monitoring Techniques

- A = Absolute means, i.e. Faraday cup, nuclear emulsions, counter telescope, etc.
- R(660) Pertains only to Russian work and means, relative to the cross section value obtained at 660 Mev, which in turn was measured absolutely.

R(Na²⁴) = Relative to the Al²⁷(p,3pn)Na²⁴ excitation function. Activity Measurement Techniques

- B = Beta counting undefined.
- B(E) = Beta external, i.e. Geiger-Müller, end window proportional counter, etc.
- $B(I) = Beta internal, i.e. 4\pi, 2\pi$, or Internal gas counter. G = Gamma measurement.

Remarks

- a = Aamodt et al. Values corrected according to Crandall et al.⁽³⁹⁾
- b = Extrapolated values from Crandall et al. (used by Hicks et al.⁽⁵⁹⁾.
- c = Results normalized to the value of Crandall et al. at 350 Mev.
- d Results by Burcham et al.⁽⁴⁶⁾ reviewed by Rosenfeld et al.⁽⁴²⁾ for their correctness, and partly corrected by Symonds et al.^(46a).
- e Part of these results were measured independently by other groups. They were all corrected for backscattering coefficient, as reported in Ref. 45.

f = Value quoted in Ref. 49.

Figure 1

EXCITATION FUNCTION OF THE REACTION C¹²(p,pn)C¹¹

FROM THRESHOLD TO 6 BEV

----- Combined excitation function of Aamodt et al.⁽³⁶⁾ (corrected) and Crandall et al.⁽³⁹⁾

- - - Best continuation proposed, on the basis of literature data at higher energies.



23a

2. Al²⁷(p,3pn)Na²⁴

The first full report about the excitation function of this reaction was given by $Hintz^{(51)}$. He used the 'stacked foil' technique and took special care to collimate the proton beam in order to reduce the energy spread in his stack of foils. (He found that the energy spread in a stack of foils increases as E^{-1}). The beam was monitored by the $C^{12}(p,pn)C^{11}$ reaction, and he used the cross-section values of Aamodt et al. (36). The activity was measured in all cases by beta-measurement techniques but, despite the care he took in considering all possible sources of error in the evaluation of his results, he did not take into account the difference in the backscattering coefficients later reported by Seliger , nor did he determine any selfabsorption correction factors for his positron emitters. Therefore his results were high. However, he took care of the neutron background and possible proton losses that might occur in the stack. Later, Hintz and Ramsey (35) redetermined the above excitation function, but they conducted their experiment in exactly the same way, most attention being given to the redetermination of the $Al^{27}(p, 3p3n)Na^{22}$ reaction excitation function relative to that of the $Al^{27}(p,3pn)Na^{24}$. The excitation function for Na²⁴ production did not change. In both cases their results were reported in graphical form and no numerical values were given. Rosenfeld et al. (42)argued that the results of Hintz and Ramsey were high only by the same factor as those of Aamodt et al. (36), i.e. 13%.

Nevertheless, they pointed out that, although in this reaction the neutron background contribution is less significant because of its high threshold, the 'stacked foil' technique should be used with extreme care, especially near the threshold.

Hintz, as well as Hintz and Ramsey, calculated the threshold for the $Al^{27}(p,3pn)Na^{24}$ reaction and found it to be 32 Mev on an energy basis (-Q) alone. If the Coulomb barrier is taken into consideration, then the threshold rises to 44 Mev. However, both $Al^{27}(p,3pn)Na^{24}$ and $Al^{27}(p,3p3n)Na^{22}$ reactions seem to have much lower thresholds. This was attributed to emission of heavier nuclides (deuterons, alpha particles, Li) rather than individual nucleons in the low energy region, in agreement with evidence accumulated by other investigators^(52,53,54,55).

Marquez and Perlman⁽¹²⁾ reported a value of 10.2 mb at 335 Mev, in agreement with the value of 10 mb given by Hicks, Stevenson, and Folger⁽⁵⁶⁾ at 340 Mev. Later Marquez⁽⁵⁷⁾ reported a value of 10.8 mb at 420 Mev. The beam intensity was monitored by absolute means, and therefore the latest value by Marquez (at 420 Mev) has been accepted as one of the most accurate values in the literature \sqrt{R} osenfeld et al.⁽⁴²⁾7.

The excitation function of Hicks, Stevenson, and Folger (56) was not found in the literature, but some of their values were quoted in various papers (58,67).

Further work in the low energy region was reported by Hicks, Stevenson, and Nervik⁽⁵⁹⁾ for the energy range between 32 and 340 Mev. They used thin targets in order to avoid neutron contribution and based their results on Crandall's cross sections (39) for the $C^{12}(p,pn)C^{11}$ reaction, which they used as a monitor. In addition to their relative measurements, they conducted two independent measurements in which the beam was monitored by a Faraday cup, one at 32 Mev using a linear accelerator, and one at 340 Mev in the Berkeley Synchrocyclotron.

In the high energy region, their results agreed with those of Marquez⁽⁵⁷⁾ and Crandall et al.⁽³⁹⁾within experimental error. In the lower energy region, however, their results were lower than those of Hintz and Ramsey⁽³⁵⁾ even when the latter were corrected for the monitor values.

Work on the excitation function of the $Al^{27}(p,3pn)Na^{24}$ reaction was mainly concentrated in the high energy region (39,44,47,48,60,61,62,63,64).

Table II is a summary of all cross section values found in the literature. The results of Hintz and Ramsey are not included in this table, but their excitation function reported in the literature, reduced by 13%, is shown in Fig. 2, where all these results are plotted. (See pp. 27 - 30.)

3. $\underline{Cu^{65}(p,pn)Cu^{64}}$

Meadows⁽⁶⁵⁾ gave the first excitation function for the formation of Cu^{64} in the low energy region up to 100 Mev, as part of his study of the spallation of Cu by protons. He monitored the proton beam by means of the $\operatorname{Al}^{27}(p,3pn)\operatorname{Na}^{24}$ reaction, using the cross section values given by Hintz and

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COLLECTED CROSS SECTION VALUES FOR THE

A1²⁷(p,3pn)Na²⁴ REACTION

Energy in	Beam Monitoring	Activity Measurement	Reported value		
Mev	Techniques	Techniques	in mb	Ref	Remarks
32	A	B(E)	0.005	59	8
50	R(C)	B(E)	1.5	59	
60	R(C)	B(E)	5.4	59	
70	R(C)	B(E)	8.2	59	
80	R(C)	B(E)	10.3	59	
82	R(C)	B & G	10.9 ± 0.4	63	
90	R(C)	B(E)	10.7	59	
90			9.2	56	Ъ
110	R(C)	B(E)	10.6	59	
110	R(C)	B & G	10.9 ± 0.4	63	
125	R(C)	B(E)	10.4	59	
134	R(C)	В & С	10.9 ± 0.4	63	
135	R(C)	B(E)	9.7	59	
150	R(C)	B(E)	9.3	59	
150	R(660)	В	12.0 ± 0.3	47	c
168	R(C)	B & G	10.1 ± 0.3	63	
175	R(C)	B(E)	8.9	59	
190			9.2	56	b
196	R(C)	B & G	9.92 ± 0.3	63	
200	R(C)	B(E)	8.6	62	đ
200	R(C)	B(E)	9.0 & 9.3	59	
202	R(C)	Ğ	9.3 ± 0.6	44	e
225	R(C)	B & G	10.1 ± 0.3	63	
225	R(C)	B(E)	10.4	59	
250	R(C)	B(E)	9.9	59	
259	R(C)	G	9.7 ± 0.5	44	6
260	R(C)	B(E)	9.5	62	đ
260	R(660)	В	11.3 ± 0.3	47	c
263	R(C)	B & G	11.2 ± 0.3	63	
294	R(C)	G	9.5 ± 0.4	44	e
300	R(C)	B(E)	11.2	59	
300	R(C)	B(E)	10.0	62	đ
325	R(C)	B(E)	11.3	59	
330	R(C)	B & G	11.7 ± 0.4	63	

Table II (Contd.)

Energy	Beam	Activity	or Descented melling		
in Mev	Techniques	Techniques	in mb	Ref	Remarks
335	A	B(E)	10.2	12	
340	R(C)	B(E)	11.5	59	
340			10.0	56	Quoted in Ref. 58
342	R(C)	G	10.0 ± 0.5	44	0
350	A	B(I)	11.1 ± 0.2	39	
350	A	B(E)	11.1 & 11.3	59	
350	R(C)	B(E)	10.2	62	đ
380	R(C)	B(E)	10.3	62	Based on carbon value from Ref. 45
420	A	B(E)	10.8 ± 1	57	
4 2 6	R(C)	B & G	11.9 ± 0.4	63	
450	R(660)	В	11.2 ± 0.4	47	с
470	R(C)	B(E)	10.7	62	Based on carbon value from Ref. 42
56 0	R(660)	В	10.7	47	c
590	A	B(E)	11.0 ± 0.1	64	
600	R(C)	B(E)	10.0	62	Based on carbon value from Ref. 45
600	A	В	11.0	61	f
650	R(C)	B(E)	10.9	62	Based on carbon value from Ref. 47
66 0	A	В	11.00	47	C
8 30	R(C)	B(E)	10.0	62	Based on carbon value from Ref.46a
980	R(C)	B(E)	10.2	62	Based on carbon value from Ref. 45
1000	A	B (E)	10.1	61	f
1600	A	B(E)	8.7	61	f
2200	A	B(E)	8.8	61	f
2200	A	B(E)	9.0	68	
3000	A	B(E)	8.1	61	f

Table II - Explanation of Symbols

Beam Monitoring Techniques

- A = Absolute methods, i.e. Faraday cup, nuclear emulsions or other specified techniques.
- R(660) = Relative to the cross-section value at 660 Mev.
 - R(C) = Relative to the C¹²(p,pn)C¹¹ excitation function.

Activity Measurement Techniques

- B = Beta measurement technique unspecified.
- B(E) = Beta external, i.e. Geiger-Müller end window proportional counters or others.
- $B(I) = Beta internal, i.e. 4\pi, 2\pi, or Gas counter.$
 - G = Gamma measurement techniques.

Remarks

- a = Quoted by Hicks et al. (59) as taken independently by
 S. Gilbert by absolute means.
- b = Values quoted in Ref. 67, but not found in the literature.
- c The results were calculated by normalizing the value obtained at 350 Mev to the σ (Na²⁴) value reported by Crandall et al.⁽³⁹⁾.
- d = Results calculated from a reported graph of the $\overline{O}_{(C^{11})}/\overline{O}_{(Na^{24})}$ ratio based on Crandall et al. excitation function up to 350 Mev and, for the remainder, on the nearest dependable value reported by various groups, as indicated.
- e = Results calculated whenever possible on the basis of \mathcal{O}_{C11} values obtained by the same author $^{(44)}$, otherwise the values of Crandall et al. $^{(39)}$ were used.
EXCITATION FUNCTION OF THE REACTION <u>A1²⁷(p,3pn)Na²⁴</u> <u>FROM THRESHOLD TO 3 BEV</u>

Data of:
Hintz and Ramsey⁽³⁵⁾ reduced by 13%⁽³⁹⁾
Hicks, Stevenson, and Nervik⁽⁵⁹⁾
Hicks, Stevenson, and Nervik⁽⁵⁹⁾
Yule and Turkevich⁽⁶³⁾
Friedlander, Hudis, and Wolfgang⁽⁶¹⁾
Prokoshkin and Tiapkin⁽⁴⁷⁾
Marquez⁽⁵⁷⁾
Goebel and Schultes⁽⁶⁴⁾



Ramsey⁽³⁵⁾. He also used the 'stacked foil' technique and measured the activity of the spallation products by a Geiger-Müller end-window counter, but did not take into account the backscattering effects reported by Seliger⁽⁴¹⁾ at all. Therefore his results should be high on account of three sources of error.

- (a) The use of the Hintz and Ramsey results which, as already mentioned, were high by at least 13% (P. 24).
- (b) The neglect of the difference in the backscattering coefficients for positrons and negatrons⁽⁴¹⁾.
- (c) The contribution of the secondary neutron reaction $Cu^{65}(n,2n)Cu^{64}$. This reaction has a cross section of about 1 barn for 15 Mev neutrons^(53,54,66) while, for higher energy neutrons, it falls rapidly to reach 98 mb for 90 Mev neutrons⁽⁶⁷⁾. Hence the effect of secondary reactions, although non-significant for the first foils of the stack, becomes increasingly important in the lower energy region, since the neutron flux increases and the cross section of the (n,2n) reaction becomes higher.

Indeed, Meadow's results were found to be higher than those of Coleman and Tewes⁽⁶⁷⁾ and Yule and Turkevich⁽⁶³⁾ at comparable energies. However, the results of Meadows in the 100 Mev region, when reduced by 15%, agree within experimental error with those of Coleman and Tewes. As far as the results of Yule and Turkevich are concerned, they are definitely low in the lower energy region, the error resulting from the monitor used. They monitored the $Cu^{65}(p,pn)Cu^{64}$ reaction with that of $Al^{27}(p,3pn)Na^{24}$ which they in turn determined with respect to $C^{12}(p,pn)C^{11}$. Their cross section values for the $Al^{27}(p,3pn)Na^{24}$ reaction are definitely low in the region of 80 - 120 Mev while, above this energy, their results agree well with the generally accepted values in the literature. (See Table II and Fig. 2.) Hence the cross section values for the $Cu^{65}(p,pn)Cu^{64}$ reaction in the lower energy region are low, while the values in the higher energy region are in fair agreement with those reported by other investigators (10,68,69,70,71).

Cohen et al. (72) have also reported an excitation function for the $Cu^{65}(p,pn)Cu^{64}$ reaction measured with respect to the $Cu^{63}(p,n)Zn^{63}$ reaction. No numerical values are given but, from the reported graph, a cross section of 660 mb was deduced at 24 Mev. This value seems to be very high, although the associated error given by the authors, ± 25%, may lower it to about 500 mb, thus bringing it in fair agreement with the low energy region results of Meadows. It was not possible to assess the reason for this unusually high value. It is felt that this may be due to the high reference cross section used for the $Cu^{63}(p,n)Zn^{63}$ reaction. They used a value of 530 mb at 13 Mev, to which they normalized their results. However, this value is 25% higher than the corresponding value found by Meadows for the same reaction at the same energy, measured with respect to the cross sections of Hintz and Ramsey⁽³⁵⁾ for the Al²⁷(p,3pn)Na²⁴

reaction. It is also possible that their activity measurement techniques are partly responsible for the error.

Table III is a summary of all values reported in the literature for the $Cu^{65}(p,pn)Cu^{64}$ reaction cross section. Part of this table was taken from Yule and Turkevich⁽⁶³⁾. Fig. 3 is a plot of all tabulated data. The lower energy portion is the excitation function of Meadows uncorrected. (For Table III and Fig. 3, see pp. 34 - 36.)

VII SCOPE OF THE PRESENT WORK

The Al²⁷(p,3pn)Na²⁴ and Cu⁶⁵(p,pn)Cu⁶⁴ reactions are extensively used as monitors.

Lack of agreement in the cross section values obtained in the low energy region, due to errors already discussed, as well as lack of correlation between these reactions on a common basis define the scope of the present work.

The excitation functions of the above reactions were studied in the energy region of the McGill Synchrocyclotron, i.e. from their threshold up to 90 Mev. The reaction $C^{12}(p,pn)C^{11}$ was used as a monitor in order to provide a common basis for the correlation of these reactions.

Improved beta and gamma measurement techniques were used to determine the variation in the cross section values obtained from each type of measurement.

Table	III

	COLLECTEI	CROSS	SECTI	ON VALUE:	5 FOR	THE	
	<u>(</u>)u ⁶⁵ (p,p	on)Cu ⁶⁴	4 REACTI	ON		
gу	Beam Monitoring	Activ Measure	ity ment	Reported	value)	

Energy in Mev	Beam Monitoring Techniques	Activity Measurement Techniques	Reported value in mb	Ref	Remarks
80	$R(Na^{24})$	B(E)	200	63	$\begin{array}{c} a \\ Based \\ H & R \end{array} \begin{pmatrix} on \\ 35 \end{pmatrix}$
82	Ħ	B & G	108 ± 4.2	63	Ъ
90	đ	B(E)	190	65	a Based on H & R(35)
90	Ħ	B(E)	126	67	8.
100	Ħ	B(E)	150	65	a Based on H& R(35)
110	14	B & G	93.0 ± 3.7	63	b
134	特	B & G	74.6 ± 2.9	63	Ъ
168	Ħ	B & G	65.8 ± 2.6	63	b
190	8	B(E)	77	67	a
196	Ħ	B & G	64.3 ± 2.5	63	Ъ
225	Ħ	B & G	57.9 ± 2.3	63	b
263	Ħ	B & G	55.0 ± 2.1	63	Ъ
280	12	B & G	69	70	C
330	Ħ	B & G	55.9 ± 2.2	63	b
340	A		73	10	8.
380	$R(Na^{24})$	B & G	68 , 83	70	c
400	17	B & G	67, 73, 71	70	C
426	Ħ	B & G	51.6 ± 2.0	63	Ъ
485	-		71	71	a
600	$R(Na^{24})$	B & G	51	70	с
800	Ħ	B & G	60	70	c
1000	Ħ	B & G	56	70	c
1300	ti	B & G	59	70	c
1600	a a a a a a a a a a a a a a a a a a a	B & G	62 .	70	c
2200	Ħ	B & G	58,65	70	c
2600	11	B & G	54	70	c
2900	11	B & G	58	70	c
3000	ti	B & G	62, 71, 73	70	c

Table III - Explanation of Symbols

Beam Monitoring Technique

A = Absolute methods specified in the quoted references. $R(Na^{24}) = Relative to the Al^{27}(p, 3pn)Na^{24}$ excitation function values.

Activity Measurement Techniques

- B(E) = Beta external, i.e. Geiger-Müller end window proportional counters or others.
- B & G = Beta and gamma measurement techniques used either independently or after intercalibration.

Remarks

- a = Values quoted by Yule and Turkevich⁽⁶³⁾ in their literature review.
- b = Results reported by Yule and Turkevich⁽⁶³⁾ based on their $Al^{27}(p,3pn)Na^{24}$ excitation function which, as indicated in Table II, was determined relative to the $C^{12}(p,pn)C^{11}$ cross-section values reported by Crandall et al.⁽³⁹⁾.
- c = The value of $\mathcal{O}_{(Na^{24})}$ taken as 10.7 ± 0.6 throughout the whole range of energies studied. In a

private communication, Dr. Friedlander stated that a reassessment of the values reported in this paper led to better agreement with those of Yule and Turkevich.

EXCITATION FUNCTION OF THE REACTION <u>Cu⁶⁵(p,pn)Cu⁶⁴</u> FROM THRESHOLD TO 3 BEV

----O--- Excitation function of Meadows⁽⁶⁵⁾ (uncorrected).

- - - Excitation function at high energies, based on literature data.

• Yule and Turkevich⁽⁶³⁾

Coleman and Tewes (67)

△ Markowitz, Rowland, & Friedlander⁽⁷⁰⁾



EXPERIMENTAL TECHNIQUES

I IRRADIATIONS

All irradiations were performed in the circulating beam of the McGill Synchrocyclotron. The energy of bombardment was varied by inserting the target at various distances from the centre of the cyclotron. Kirkaldy's (73) standard curve of energy variation versus radial distance of the beam was used to determine the energy of the bombardment. This curve, corrected for radial oscillations (74), is shown in Fig. 4. The energy spread of the beam was ± 2 Mev.

The length of irradiation was varied from 15 minutes to 1 hour according to the energy of bombardment, so that production of sufficient activity was assured in every case.

II TARGET ASSEMBLY

1. Material

The target consisted of a stack of three foils, one for each reaction studied. These foils were arranged in the following sequence; aluminium, carbon, and copper, the proton beam impinging first on the aluminium. A check for recoil losses, carried out by Mr. C.L. $\operatorname{Rao}^{(75)}$ in this Laboratory, proved that they are insignificant for the energy range of interest, at least for the thickness of the target materials used. Hence, no guard foils were necessary.

The aluminium foil used had a purity greater than 99.9% and was 0.002" thick. The uniformity of the thickness

ENERGY CALIBRATION CURVE FOR THE

McGILL SYNCHROCYCLOTRON



was checked by a micrometer, and it was found that deviations never exceeded 0.0001[#]. The superficial density of this foil was also measured as an additional check by cutting areas of various sizes and weighing them. The superficial density was $13.30 \pm 0.15 \text{ mg/cm}^2$.

The copper foil was obtained from Johnson, Matthey and Co., Limited, 73-83, Hatton Garden, London, E.C. 1, England. According to the Company's specifications, it was 99.99% pure. Its thickness was also 0.002, and the deviation never exceeded 0.0001. The superficial density, measured in exactly the same way as that for the aluminium, was found to be $45.57 \pm 1.0 \text{ mg/cm}^2$.

The carbon source presented some difficulties. Preliminary experiments carried out with plastic materials, such as various types of polyethylene, polyisopropylene, teflon, etc., showed that they melted during irradiations. The problem was finally solved by using thin carbon foils prepared from fine grain, high purity graphite electrodes supplied by the McGill Radiation Laboratory. Dr. T.M. Kavanagh of that Laboratory had proposed the use and developed the technique of cutting very thin and uniform foils from these electrodes. The thickness of these foils varied between 0.004" to 0.006". The deviation from uniformity of each individual foil was consistently less than 0.0001". However, it was felt that the density of the electrodes used might vary as much as 10% due to imperfections in the manufacturing process. Therefore, as an additional precaution,

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the superficial density of these foils was checked before use. It was found to vary between 18 and 25 mg/cm^2 according to the thickness of each individual foil. The accuracy of the measurements was better than 3%.

The total target thickness, as it can be easily deduced, never exceeded 84 mg/cm^2 or 0.025 cm, i.e. it was thin enough so as not to cause attenuation of the beam intensity or degradation of its energy.

2. Preparation

A most important problem arising from internal beam bombardments, where monitors are used, is the proper alignment of the exposed area of the target stack so that all foils receive the same beam flux. Particular attention must be given to the leading edge of the target which receives most of the beam.

The problem is usually solved by cutting the stack after it has been mounted on the target holder. Thus a perfect alignment of the exposed area is obtained. The same technique was applied in this work. A target cutting device, shown in Fig. 5, was constructed which allowed outting of the three exposed edges of the target stack as well as a close alignment of the fourth edge. The target was cut by a sharp surgical scalpel because the brittleness of the carbon foil did not allow use of shearing instruments. After being cut, the target was transferred to the target holder by means of the detachable cutting lead which also served to hold the foils together. The leading edge was then freed by

TARGET CUTTING DEVICE AND STEPWISE

PROCEDURE FOR TARGET PREPARATION

TARGET CUTTING DEVICE



MOUNTING PROCEDURE





Place "leading edge clamp" and cut along for the leading edge ot target stack.



Remove "pressure block" and detach "mounting block" without releasing target.



Mount target on "target holder", then release it from "leading edge clamp" loosening the screws of the cutting lead. An excellent alignment of the exposed area was thus obtained.

III TARGET CHEMISTRY

1. Aluminium

The radioactive nuclides, Na²⁴ (t₁ = 14.9 hrs) and Na²² (t₁ = 2.58 yrs) have half-lives long enough so that no interference occurs from all other radioactive nuclides likely to be produced from the bombardment of aluminium. Even F^{18} (t₁ = 1.87 hrs), which is formed at higher energies, will not interfere if the activity measurement is started 15 hours after bombardment. Usually the measurement of Na²⁴ was started about 24 hours after the end of bombardment and the F^{18} had died out completely. Hence the activities of Na²⁴ and Na²² could be measured easily, even without chemical separation. In fact, simply dissolving the aluminium and preparing the necessary sources for beta and gamma measurements would suffice. Of course the beta measurement would be slightly affected by the presence of the large amount of AlCl₃ and corrections for the absorption of radiation by the source would have to be applied. Another factor that might also affect the beta measurements to an unknown extent would be the deliquescence of the Al salt.

In order to avoid all those possible sources of error, it was decided to separate the Na²⁴ from the aluminium matrix. This separation was readily achieved by ion exchange. The Al target was dissolved in a small volume of HCl to which a few drops of H_2O_2 were added to enhance the action of HCl. One mg of NaCl carrier was added to the mixture in order to minimize possible losses of Na²⁴ due to exchange with the glassware or by adsorption. The mixture was heated gently to destroy the excess H₂O₂ and to eliminate the Cl₂ gas formed by the action of the H_2O_2 on the HCl, and then taken to dryness by gentle evaporation under an infrared lamp to prevent losses by bumping of the solid salts towards the end of the evaporation. The residue was redissolved in as little dilute HCl (about 3 N) as possible. The solution was then adsorbed on a Dowex-50 cation exchange column (100 -200 mesh) 12 cm high and 0.6 cm in diameter. The column was pretreated with about 50 cc of concentrated HCl (12 N), washed with water, and conditioned by passing about 50 cc of After the AlCl₃ and NaCl mixture was adsorbed, 0.7 N HC1. the column was eluted with 0.7 N HCl. The flow rate was adjusted to about 1.3 ml/min. The elution was stopped after 70 cc had passed. A diagram of the elution process is shown in Fig. 6. The only cation the eluate contained was Na. The aluminium started to appear after about 800 ml of eluate had passed. The eluate was then taken to dryness by gentle evaporation under an infrared lamp, dissolved in H₂O and transferred quantitatively into a volumetric flask, made to volume, and suitable aliquots taken for the activity measurement.

The method just described is a simplification of a previous one developed by Hollbach and $\text{Yaffe}^{(76)}$. The

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ELUTION CURVE FOR Na SEPARATION

FROM A1

Resin used: Dowex 50

Eluant - 0.7 N HCl



technique is the same. The difference, however, lies in the use of a better type of resin, smaller size of column and elimination of the purification step they had to take in order to remove resin impurities from their eluate.

The efficiency of the elution process was determined by using a known amount of Na²² activity measured on a 4π counter, conducting a separation as described above, and determining the amount of Na²² activity recovered on the same counter.

Table IV shows the results obtained during a series of such experiments. The average efficiency calculated from these results was 99 - 0.5%.

Table IV

EFFICIENCY DETERMINATION OF Na SEPARATION BY A DOWEX-50 CATION EXCHANGE COLUMN

Elution No.	Activity in cpm	Activity out cpm	% Efficiency
1	1.241×10^6	1.223 x 10 ⁶	98.53
2	1.198 x 10 ⁶	1.189 x 10 ⁶	99.21
3	1.165 x 10 ⁶	1.157 x 10 ⁶	99 • 3 5
4	1.134 x 10 ⁶	1.117 x 10 ⁶	98.43
5	1.094 x 10 ⁶	1.085 x 10 ⁶	99.18

Column dimensions: h = 12 cmd = 0.6 cm

Average: 98.94 ± 0.5

By careful application of the above technique, carrier-free Na activity may be obtained with equally high efficiency. No chemical yield determinations were necessary for the determination of the amount of recovered activity. As an additional precaution, the elution was never stopped before it was made sure that no activity remained in the column.

2. Copper

Copper was chemically treated in order to separate the Cu isotopes from other concurrent activity, mainly that of Zn, Co, and Ni, which interfere with the measurement of Cu^{64} . No carrier was necessary, as the target itself acted as a carrier for the Cu activity. The ion exchange technique was used in this case too.

The method employed was that described by Kraus and Moore(77). The copper foil was dissolved in a small amount of concentrated HCl to which a few drops of H202 were added. The solution was heated to destroy the excess H_2O_2 and to eliminate the Cl_2 gas formed by the action of H_2O_2 on the HCl, taken to dryness and the residue redissolved in a small amount of concentrated HCl. The new solution was adsorbed on a Dowex 1 anion exchange column (200 - 400 mesh), preconditioned by passing some 20 ml of 4.5 N HCl acid, and then the elution step followed. The passage of 4.5 N HCl acid carried away Co and Ni contaminations while the copper layer moved slowly towards the bottom of the column. The strong yellow-green colour of the copper layer facilitated

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the inspection of the separation and rendered unnecessary the use of exact column dimensions, provided the resin was adequate to adsorb all the copper ions. The column used was 12 cm high and 0.6 cm in diameter (as opposed to 29 cm high and 0.29 cm² in area used by Kraus and Moore). When the copper was about to come out, the eluant was changed to 1.5 N HCl acid which brought out the copper within the next 5 - 10 ml Zinc was retained on the column. Part of the of eluate. head of the eluate as well as part of the tail were rejected so that the purity of the copper fraction could be ensured. The eluate was received directly into a volumetric flask, made to exact volume, and suitable aliquots taken for the activity measurements. The fraction of the copper activity recovered was determined colorimetrically by the diethyldithiocarbamate method (78), slightly modified by G.V.S. Rayudu⁽⁷⁹⁾ to suit the needs of this Laboratory. The absorbance of the copper diethyldithiocarbamate complex was measured by a model DU Beckman spectrophotometer in 1 cm Corex cells at a wavelength of 430 mm. The calibration curve of absorbance versus amount of CuO is shown in Fig. 7.

The elution yields for Cu ranged from 30 - 90%. In the cases of higher yields, some Co contamination was noticed and resolution of the decay curves was necessary.

3. Carbon

If only one method of activity measurement were to be used, chemical treatment of carbon would not be necessary from the point of view of activity contamination. C¹¹ is the only measurable activity produced by bombardment of

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STANDARD ABSORBANCE CURVE FOR COPPER

DIETHYLDITHIOCARBAMATE COMPLEX



carbon with protons. In this work, gas phase as well as gamma measurement techniques were employed, and therefore the chemical treatment of the carbon target was necessary.

(a) Combustion

Carbon was converted to CO, by a combustion method suggested by Dr. R.A. Sharp of Brookhaven National Laboratory (80). The apparatus used was also suggested by him and is shown in Fig. 8. The method, a modification of combustion techniques developed by other investigators in connection with gas phase measurement of C14 (81,82,83,84,85,86), was designed for use in spallation studies at high energy, where C¹¹ is formed in rather low yields as a spallation product. Although conditions in the present work allowed some simplifications in the procedure as well as in the construction of the apparatus, it was thought desirable to construct the combustion apparatus exactly as indicated by Dr. Sharp in view of possible future use in spallation investigations. Simplifications of the procedure will be indicated along with a description of the method.

The apparatus was preconditioned by over-night evacuation and flushing with O₂ before irradiation was started.

The gas counter must be isolated from the system before flushing it to prevent contact of the counter tube with O_2 . The PbO₂-CuO trap (PC) was set to 350°C and the NiO furnace (NF) to 950°C. After irradiation the target was weighed, placed into the target solution flask (TF) and

CARBON COMBUSTION APPARATUS

```
CG:
          Carrier gas cell
     TF:
          Target solution flask
     FF:
          Flask filling funnel
     CC:
          Condenser
          Sulfuric acid scrubbing tube
     ST:
          PbO<sub>2</sub>-CuO trap (operated at 350°C)
     PC:
          NiO furnace (operated at 950°C)
     NF:
                            ( I - For the purification of
          ( the CO_2 if necessary
Ba(OH)<sub>2</sub> bubblers (II - For the preparation of
     BT:
                             BaCO<sub>3</sub> sources
          Bubbler filling funnel
     BF:
          Calibrated needle valve
      N:
          Calibrated flow meter (maximum flow - 100 ml/min)
     FM:
          Calibrated manometer
     MT:
     PG:
          Electronic Gauge (Phillips or Pirani)
     CT:
          Gas counting tube
          Barostat (filled with DC704 to a height of about
     BS:
                                                       12 cm)
          -30" to +15" Bourdon gauge
     GA:
          Liquid air trap for the diffusion pump
     DT:
     DP:
          Diffusion pump
          Air filter
     AF:
     FP:
          Fore pump
 TRAP I:
          Immersed in dry ice-acetone mixture
TRAP II:
          Immersed in liquid nitrogen or liquid air
          Stopcocks ( One way
1 - 29:
          Medium porosity sintered glass plugs
        Conical ground glass joints
      :
          Ball and socket joints (for better flexibility)
      = :
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50a

attached to the system. The target solution section was then evacuated and partly refilled with 0_2 , or with 0_2 and 002. The latter was enclosed in the carrier gas cell (CG) whenever a need for it was anticipated.* The Van Slyke-Folch chromic acid combustion fluid (83, 84) was added next through the filling funnel (FF) and the mixture boiled for about one minute while 0, was bubbled through it.** The volatile products of the combustion, swept by the 0_2 , were passed through the condenser (CC) and the sulphuric acid trap (ST) which was filled with 80% H₂SO₄ to remove SO₂ produced during The gases were then passed through the PbO2 combustion. CuO trap where H₂ and nitrogen oxides were removed.*** Next, the gases were introduced into the NiO furnace (NF)****, where all products carried by the sweeping gas were catalytically converted into CO_2 . The mixture of CO_2 and O_2 was then passed through the two radiator-type traps, bypassing the

- *The use of CO₂ carrier is necessary only in spallation studies where the C¹¹ produced is very little No such need ever arose in the present work.
- **If metal targets are used, dissolving them before adding the combustion fluid is recommended.
- ***Nitrogen oxides are usually produced when certain targets, mainly in spallation studies, are dissolved in HNO3 before combustion. They are severe poisons for gas counters and must be removed.
- ****The NiO catalyst is made of nickel chips introduced into the furnace and oxidized in situ by passing O₂ at a temperature of 700°C or above.

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Ba(OH), bubbler (BTI).*

In trap I, immersed in dry ice-acetone, water vapours were removed while in trap II, immersed in liquid air, CO_2 was frozen and thus separated from the O_2 . The system was swept for about 5 minutes at constant rate, controlled by a calibrated needle valve (N) and a flowmeter (FM). Then the oxygen supply was cut off and pumping continued until a vacuum of about 26^m, read on a Bourdon Gauge (GA), was reached. The flow rate was kept constant by gradually opening the needle valve.

The CO_2 in trap II was then pumped free of oxygen after cutting off the pump line, trap I, and the combustion section. A vacuum of 10^{-4} to 10^{-5} mm Hg was recommended by Dr. Sharp in order to ensure absence of impurities, mainly water vapours, which affect the operating characteristics of the counter. This vacuum was never achieved in this work. Instead, the counter was carefully flushed with methane before CO_2 was distilled into it.^{**}

After flushing and evacuating to a pressure of 10^{-3} mm Hg, the CO₂ was allowed to thaw and expand into the volume

*The purpose of the Ba(OH)₂ bubbler (BT) is to separate CO₂ in cases where contaminants, not separable by easy trapping schemes, are present in the combustion gases. Such is usually the case in spallation of high Z elements where Rn and Xe are found among the spallation products. CO₂ can be regenerated by acidifying the bubbler solution,

**In spite of the careful flushing, it appears that impurities, mainly due to the CH₄ gas, were unavoidably introduced into the counter which caused a delay in the stabilization of the counter characteristics. This effect is discussed in a later section of this work.

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bound by stopcocks 13, $14\sqrt{15}$, 15, 16, 18, 19, 21, and 22. The amount of CO₂ obtained was determined by reading the pressure on a calibrated mercury manometer (MT). * The CO₂ could then be either quantitatively transferred into the gas counter (CT) by immersing the bottom into liquid air, or allowing it to In the latter case, the expand into the gas counter volume. aliquot of CO₂ introduced could be readily calculated from the manometric pressure, if the volume of the counter were known, by application of the ideal gas law. The validity of the ideal gas law was checked by similar calculations, using the Van der Waals and the virial equations. The agreement was better than 0.2% when applied to a pressure of 100 mm Hg.

After the gas counter aliquot was secured, the remainder of the CO_2 was swept by means of methane gas through a $Ba(OH)_2$ trap (BT II), added to the line for the purpose of obtaining a second aliquot to be used for gamma radiation measurements. The necessary precautions to prevent contact of $Ba(OH)_2$ with the atmosphere were taken. The $BaCO_3$, formed in trap BT II, was quickly filtered under suction followed by ten successive washings with degassed distilled water so that all excess $Ba(OH)_2$ would be removed. The precipitate was then washed further with ethanol and ether, sucked dry, and introduced into a screw cap vial for gamma activity measurements.

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^{*}The manometer was calibrated by combusting weighed amounts of benzoic acid and plotting the expected amount of CO₂ versus the manometric pressure. Fig. 9 shows the calibration curve obtained. The calibration was also checked by liberating CO₂ from known amounts of Na₂CO₃. The agreement was excellent.

CALIBRATION CURVE FOR THE MANOMETER

OF THE C COMBUSTION APPARATUS



After all the CO_2 was removed from the system, methane was admitted into the gas counter to bring the pressure to one atmosphere. The counter was then removed from the line, and the CO_2 enclosed was measured through its beta radiation.

(b) Analysis of the samples

The volume of the gas counter was measured by the toluene method (86). The tube was filled with reagent grade redistilled toluene and weighed before and after filling. The specific gravity of the toluene at the temperature of the experiment was obtained from literature data (87). The experiment was repeated in exactly the same way using benzene as a filling liquid. The results agreed perfectly. The average of the two results was 99.65 \pm 0.05 ml.

No analysis was necessary for the gas counter aliquot.

The BaCO₃ sample was analysed volumetrically after all C¹¹ had decayed away. BaCO₃ was dissolved in an excess of O.1 N HCl acid added through a calibrated micro-burette under continuous stirring. A few drops of bromothymol blue indicator were added and the excess HCl titrated with O.1 N KOH, also added through another calibrated micro-burette. A sharp colour change from yellow to blue indicated the end of the titration. The accuracy of the method, checked by repeated analyses of known amounts of BaCO₃, was better than 2%. The stability of the HCl and KOH solutions was checked periodically and before any series of analyses.

IV RADIATION MEASUREMENT TECHNIQUES

Nuclear radiations, both corpuscular (α , β^{-} , β^{+}) and electromagnetic (γ , X-rays), are detected through their interaction with matter. When radiations pass through matter, they lose their energy chiefly by interaction with electrons, which lead to dissociation of molecules or excitation or ionization of atoms.

All radiation detection devices are based on the effects of these interactions, and special detectors are used for each type of radiation, according to the efficiency with which the particular interaction occurs. The latter depends largely on the nature and energy of the radiation. Thus, corpuscular radiation ($\propto \text{ or } \beta^{t}$) is best detected through the ionization effect it produces in gases. Electromagnetic radiations are not that efficiently detected by the same effect, as the ionization they produce in gases is less intense due to their penetrability. The most efficient way to detect electromagnetic radiations is by means of the scintillations they produce in certain crystalline materials called 'phosphors'.

Of the nuclides studied, Na²⁴ decays 100% by negatron emission to several excited states of Mg^{24} which, in turn, reaches its ground state by emission of a number of gamma rays. Hence the beta decay of Na²⁴ is accompanied by gamma radiation. Cu⁶⁴ decays 19% by positron emission, 38% by negatron, and 43% by electron capture. The negatron decay leads to the ground state of Zn^{64} while both positron and electron capture decays lead to the ground state of Ni 64 . A negligible percentage (0.34%) of the total decay of Cu 64 goes to an excited state of Ni 64 , giving off a gamma ray, but this is practically useless for gamma radiation measurements. However, the positrons offer an excellent source for gamma measurement of Cu 64 .

When a positron meets a nearby electron, annihilation occurs. As a result, two gamma quanta are emitted in nearly opposite directions, each equal to 0.511 Mev. These annihilation gamma quanta accompany all cases of positron decay, and actually their detection constitutes a proof for this mode of decay.

Finally, C^{11} decays 100% by positron emission to the ground state of B^{11} . Some recent investigations⁽⁸⁸⁾ led to the discovery of some electron capture, but the percentage is insignificant (0.002%). Of course, C^{11} decay is accompanied by the annihilation radiation. The decay schemes⁽⁸⁹⁾ of these three nuclides are shown in Fig. 10.

The problem of radiation measurements for the three nuclides studied was resolved by using appropriate counting techniques. Na²⁴ and Cu⁶⁴ beta radiation was measured by a 4π gas flow proportional counter and that of C¹¹ by an internal gas counter, while the gamma radiation from all three was measured by scintillation counters.

A. Beta radiation measurement techniques

(1) 4 N counting

The theory behind beta measurement is well known
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DECAY SCHEMES OBTAINED FROM REF. 89

a. Decay scheme for C¹¹
b. Decay scheme for Na²⁴
c. Decay scheme for Cu⁶⁴



a

and need not be dealt with in the present work. Only a brief discussion of the 4π measurement technique, as developed by Pate and Yaffe⁽⁹⁰⁾ in this Laboratory, will be given.

a. Equipment

The counting equipment used was that described by (89b) them The counting chamber, shown in Fig. 11, consisted of two hemispherical brass cathodes, 7 cm in diameter, and two ring-shaped anodes made of 0.001" tungsten wire. The anodes were insulated from the cathodes by teflon insulators. The cathodes were kept at ground potential, while the anodes received a positive high potential supplied from a Nichols high voltage supply (A.E.P. 1007B). The two anodes, each acting as a separate unit of 2π geometry when the source is kept in the centre of the sphere, were connected in parallel to an Atomic Instruments preamplifier (Model 205-B). The output from this was fed into an Atomic Energy of Canada Limited amplifier discriminator (AEP 1448). The overall gain of the system was about 30,000, and a bias voltage up to 50 volts could be applied to the signal by the discriminator. Counting rates were recorded on a Marconi scaler unit (A.E.P. 908). Auxiliary equipment used included a Sola constant voltage transformer and a Lambda regulated power supply (Model 28). A block diagram of the assembled equipment is shown in Fig. 12. The counter chamber was operated in the proportional region. The counting gas was C.P. Methane at atmospheric pressure. After the sample was

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SCHEMATIC DIAGRAM OF THE

 4π counting chamber



60a



BLOCK DIAGRAM OF THE

 4π COUNTING ASSEMBLY



inserted, the counter was flushed for several minutes before starting measurements. The flow rate was controlled by a simple flowmeter attached to the line.

b. Counter characteristics

The counter characteristics were checked before work was begun. These included establishing the operating plateau region for the nuclides under study, bias curves, as well as a check of the statistical behaviour of the counter by a χ^2 test.

The plateaus were about 400 volts long and had a slope of 0.1%. A common operating voltage was selected for all nuclides at 2650 volts. The counting rates at this voltage remained constant throughout the discriminator range, indicating absence of electronic noise and good amplification of the counter signals. A setting of 12.5 volts was used on the discriminator. The χ^2 test, run with a Ra(D+E) standard under the operating voltage and discriminator settings indicated above, showed a good statistical behaviour for the counter. It was then necessary to determine correction factors for possible errors originating from the sources.

c. Pipetting errors

The sources were prepared by pipetting a small aliquot from a known volume of the solution of the nuclide to be studied, by means of a calibrated micro-pipette. In all cases duplicate 100 lambda samples were taken and mounted on thin VYNS films, prepared by the technique described by Pate and Yaffe^(90a). Sources, whose counting rates differed by more than 2%, were rejected. In most of the cases, the counting rates of the sources differed by 1% or less.

d. Dead-time losses

Although proportional counters have short resolving times, permitting the measurement of high count rates, it is necessary to know the magnitude of such losses. Pate and Yaffe^(90b) have suggested that the resolution losses are best determined empirically. A resolution loss curve, constructed by the method described by them, is shown in Fig. 13. This curve was rarely used in the present work because care was taken to keep counting rates lower than 10^5 c.p.m., where the loss is about 2.5%.

e. Source-mount absorption

The use of thin films and the high energy of the beta particles emitted by Na²⁴ (β_{max} 1.39 Mev) render source-mount absorption corrections for this particular nuclide trivial and unnecessary. The case of Cu^{64} , however, is more complicated. As already mentioned, 43% of the decay of Cu takes place by electron capture. This mode of decay is followed by emission of X-rays which in turn give rise to Auger electrons. The latter have very low energies and, although they can be detected by internal beta counting techniques, their detection probability is not always unity. That is, not all of the Auger electrons reaching the gas chamber are able to initiate an ionization event that could produce a high enough signal which could be registered as a count in the detection system.

RESOLUTION LOSS CURVE FOR THE

 4π counter



In addition to this, Auger electrons suffer great losses by absorption from the source material, as well as from the source mount. Therefore, in order to avoid introduction of such large potential sources of error, it was thought preferable to prevent them from reaching outside the source by the use of some absorber of suitable thickness. A study of the absorption of Auger electrons showed that an absorber thickness of about 110 μ g/cm² was adequate to eliminate them completely. (Fig. 14) Hence the Cu⁶⁴ sources were sandwiched between two VYNS films and the whole covered on both sides by a thin aluminium foil having a superficial density of 108 μ g/cm².

Since the source mount was not thin any more, source-mount absorption corrections had to be used for the $\beta^+(E_{max} \ 0.66 \ Mev)$ and $\beta^-(E_{max} \ 0.57 \ Mev)$ emitted by the source⁽⁸⁹⁾. The source-mount absorption corrections were obtained from the data of Pate and Yaffe^(90c) who presented a set of curves giving source-mount absorption as a function of the maximum energy of the β -emitter for various film thicknesses. A replot of these data for the particular energies of interest is shown in Fig. 15.

f. Self-absorption

This is the largest error that may be introduced into beta radiation measurements of solid sources. The

^{*}The sources were not mounted directly on the aluminium in order to prevent chemical action of the source on the Al support.

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AUGER ELECTRON ABSORPTION CURVE

O - First series of experimental points

• - Duplicate check points



SOURCE-MOUNT ABSORPTION CURVE FOR

BETA RADIATION OF Cu⁶⁴

(Data obtained from Ref. 90c)

a. (E 0.66 Mev) max

b. (E_{max} 0.57 Mev)



self-absorption effects become increasingly important as source thickness (or superficial density) increases, or the energy of the beta particles decreases. Pate and Yaffe (90c) and Yaffe and Fishman⁽⁹¹⁾ have determined the self-absorption coefficients for a large range of beta particle energies over a wide range of source thicknesses. By replotting their results, which were presented again as a set of curves giving the self-absorption as a function of the maximum energy of the beta emitter for various source thicknesses, the selfabsorption curves for the nuclides of interest were obtained, as shown in Fig. 16. From these curves, it is clearly seen that for Na²⁴ self-absorption is really unimportant for source superficial densities below 30 μ g/cm². Almost all of the Na²⁴ sources prepared in this work were less than 20 μ g/cm². (The largest sources prepared contained 2% of the total amount of the 1 mg NaCl used as carrier during separation.) Hence no self-absorption corrections were necessary for Na²⁴.

Self-absorption corrections had to be used in the case of Cu^{64} , since the large amount of Cu carrier resulted in quite thick sources, and the beta particles from Cu^{64} are much softer than those of Na²⁴.

No other corrections were applied to the counting

[•]Only in the last four irradiations was Na not separated from the Al matrix. In these cases, the sources were sandwiched between two VYNS films to prevent introduction of possible errors by the deliquescence of the AlCl₃(xH₂0). The additional VYNS films did not cause any source-mount absorption error, but self-absorption corrections were used in these four cases.

SELF-ABSORPTION CURVE FOR THE BETA

RADIATION	OF	Na^{24}	AND	Cu ⁶⁴

(Data obtained from Refs. 90e and 91)

a. Na²⁴ (β_{max}^{-} 1.39 Mev)

- b. Cu⁶⁴ (β⁺_{max} 0.66 Mev)
- c. Cu⁶⁴ (β_{max} 0.57 Mev)



69a

a. Equipment

The gas counter used in this work was that described by Bernstein and Ballentine (86). It consisted of a cylindrical glass tube about 36 cm long with an external diameter of 1.9 cm. About 30 cm of its wall was silverplated to serve as a cathode. The anode consisted of a 0.002[#] tungsten wire, taut from one end to the other along the main axis of the tube. The part of the anode that lay outside the silver-plated area was surrounded by a glass tube shield whose function was to confine the electrical field of the gas chamber within the space enclosed by the cathode only. A diagram of the construction details of the counter is shown in Fig. 17.

The counter was enclosed in a lead castle 2" thick in vertical position. The cathode of the counter was kept at ground potential, while the anode received a positive high potential from a Dynatron Radio Limited (type 200A) high voltage supply unit. The anode was connected to a cathode follower constructed in this Laboratory by J.R.S. Drouin⁽⁹²⁾. The output from this was fed into an Atomic Energy of Canada Limited amplifier-discriminator (AEP 1448). The overall The discriminator range was from 0 to 50 gain was 700. volts. Counting rates were recorded on a Marconi scaler unit (AEP 908). Auxiliary equipment included a Sola constant voltage transformer and a Lambda regulated power supply (model 25). A block diagram of the counting assembly is shown in Fig. 18.

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SCHEMATIC DIAGRAM OF THE GAS

COUNTER CHAMBER

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b. Calibration of Counter Characteristics

The counter characteristics are affected by the amount of CO_2 present in the gas filling. The calibration was divided into two main steps. In the first step, the effect of varying amounts of CO_2 on the counter operating characteristics was studied. In the second, the effect on the detection efficiency was determined.

(i) Effect of CO₂ on the Counter Operating Characteristics:

Carbon dioxide is a mild poison to any gas counting system. Bernstein and Ballentine⁽⁸⁶⁾, as well as Van Slyke et al. (93) have shown that, when the total CO_2 pressure in the counter is less than 100 mm Hg, good plateau characteristics are obtained. In fact, the recorded counts showed an increase with increasing voltage until a nearly level plateau was reached near 4000 volts. The slopes of the individual plateaus increased with increasing amounts of CO2 inside the counters, but all reached the same count at 3900 volts. The same check was done during this work at three different pressures definitely higher than those expected in the actual experiments. A mixture of $Na_2C^{14}O_3$ and $Na_2C^{12}O_3$ was prepared and the CO_2 liberated was stored into trap II of the combustion apparatus (Fig. 8). From this stock various amounts of CO2 were introduced into the counter and the plateau was determined at various

bias settings. Of course the counting rates recorded were different according to the amount of gas mixture used. The results obtained are shown in Fig. 19 (a,b,c). As it is seen, the plateau slopes did not vary much as the pressure of CO_2 increased from 52.6 to 84 mm Hg. A comparison with the plateau taken with pure CH₄ filling and an external source, also shown in Fig. 19 (d), indicates that the plateau could be improved at lower CO_2 pressures.

- (ii) Effect of CO₂ on detection efficiency:
 - Bernstein and Ballentine⁽⁸⁶⁾ as well as Van Slyke et al. (93,94) have also shown that the detection efficiency of the counter is not affected by the presence of CO₂ up to a pressure of 100 mm Hg when methane is used as the counting gas. If methane is replaced by PlO gas (90% argon and 10% methane) the efficiency falls quite rapidly when the CO_2 pressure exceeds 20 mm Hg⁽⁹⁴⁾. The effect of CO, on the efficiency of the counter was checked by plotting the plateau characteristics obtained at a fixed bias setting for the three CO2 samples previously mentioned. Fig. 20 shows these plateaus plotted as specific count rate versus operating voltage to allow for the different size of aliquots used. As it can be seen, the plateaus coincide over a voltage range from 3600

PLATEAU CHARACTERISTICS OF A GAS COUNTER

AT	VARIOUS BIAS VOLTAGES FOR
	DIFFERENT CO2 PRESSURES
8.	CO ₂ pressure - 52.6 mm Hg
b.	CO ₂ pressure - 77.8 mm Hg
c.	CO ₂ pressure - 84.0 mm Hg
d.	Pure methane filling



DETECTION EFFICIENCY OF GAS COUNTER

AT VARIOUS CO2 PRESSURES



to 3900 volts, indicating that the counting efficiency did not vary for the range of pressures used. \mathcal{A}^2 tests done in all three experiments showed that the counter operated statistically and that the CO₂ had no effect on the counter behaviour.

c. Counter efficiency determination

This operation involves the study of a number of effects (such as end, wall, radius and backscattering) which were described and extensively studied by earlier investigators (95,96,97,98) who were mainly concerned with C¹⁴O₂ gas phase counting in the Geiger-Müller region. For purposes of the present study it was not necessary to know each individual effect, but to obtain an overall efficiency factor for the particular gas counter and radiation studied. Furthermore. Bernstein and Ballentine⁽⁸⁶⁾ showed that in their type of counter, which was also operated in the proportional region, the above-mentioned effects were minimized to such an extent that a combined efficiency of 98% could be attained. According to Van Slyke et al. (93) the best way to determine the efficiency of a counter would be to measure the count of a known amount of CO₂ (yielding a pressure below 100 mm Hg) obtained from a standard. Since this work was concerned with the measurement of C^{11} rather than C^{14} , the C^{11} itself was used for the standardization of the counter. To this end, the C^{11} , obtained by proton bombardment of C^{12} , was measured by the 4π counting method and the gas counter.

The respective counting rates were converted to total absolute disintegration rates, and the ratio of the 4π results to those of the gas counter was taken. The standardization of the gas counter against the 4π had the added advantage of rendering the beta radiation measurement of C^{11} comparable to that of Na²⁴ and Cu⁶⁴. The conversion of the 4π counting rates to absolute disintegration rates is discussed in detail in Appendix I.

The overall conversion factor obtained for the gas phase measurement of C^{11} was 1.513 ± 0.069 which includes the volume ratio of the counter. The latter is a constant for any one counter and is the ratio of the volume enclosed by the cathode to its total volume. This ratio indicates the fraction of the total activity enclosed in the counter that would be ideally measured if its efficiency was 100%. This sensitive volume was measured for the particular counter in use by the toluene method (86,93) and was found to be 84.23%. If $V_{\rm R}$ is the volume ratio, F the overall factor and E the efficiency of the counter, then

$$\mathbf{F} = \frac{1}{\mathbf{V}_{\mathbf{R}} \times \mathbf{E}} \qquad \dots \qquad (5)$$

Solving for E, equation (5) becomes

$$E = \frac{1}{F \times V_R} \qquad \dots \qquad (6)$$

Since F = 1.51 and $V_R = 0.842$, the efficiency can be calculated from equation (6), i.e.

$$E = 70.8\%$$
.

This is a surprisingly low value when compared to that of 98% quoted by Bernstein and Ballentine⁽⁸⁶⁾, but true. The reasons are unknown. One plausible explanation is that the electronic equipment used may be responsible for this dis-In effect, the electronic equipment did not match crepancy. the requirements set by the above authors, as explained by Van Slyke et al. (93). The amplification gain was lower (700 instead of 1000) and the discriminator did not have the sensitivity specified. In fact, a rather high discriminator bias was used in order to eliminate the electronic noise of the system. Attempts to compensate this effect by the use of a preamplifier resulted in highly unstable operation of the counter. Only by the use of a cathode follower was reasonable stability achieved. However, it is felt that, although the efficiency of the counter was low, the stability of the counting characteristics, as already shown, makes the measurement of the C¹¹ activity reliable to within 5.7%.

d. Resolution losses

Bernstein and Ballentine $(^{86})$ reported that the gas counting system they used had a very short resolving time, permitting the measurement of very high specific activities (up to 2 x 10⁵ c.p.m.) without any appreciable resolution loss. However, from the decay curves of the C¹¹ activity measured in this work, it became apparent that the resolution losses in the gas counting system employed were quite significant even for counting rates as low as 5 x 10⁴ c.p.m. Therefore an empirical resolution loss curve was determined using the technique described by Pate and Yaffe^(90b) suitably adapted for this case.

The gas counter was filled with a mixture of CHL and CO₂ to a total pressure of one atmosphere. The partial pressure of the CO₂ was 30 mm Hg. Eight pairs of sources of increasing specific activity were prepared by evaporating some P^{32} on polyethylene strips. The sources were covered by scotch tape and attached to the outer surface of the counter. The measurement of their activities was done in the following sequence: A, A + B, B, B + C, C etc., beginning with the sources of the lowest specific activity ($\sim 3 \times 10^3$ c.p.m.). In this manner the individual sources remained fixed in the same position during counting and were removed only when they were of no further use. Thus possible errors due to geometry effects were avoided. The resolution losses, plotted as a function of apparent counting rate, are shown in Fig. 21. From this curve, the resolving time of the system was calculated by using the equation

$$N_{T} = \frac{N}{1 - N\tau} \qquad (7)$$

where N_m is the true counting rate,

N is the apparent counting rate, and

τ is the resolving time of the counting system.

At 10^5 c.p.m. apparent counting rate, the resolution loss is 1.7 x 10^4 c.p.m., that is $N_T = 1.17 \times 10^5$ c.p.m. By substituting the above values for N and N_T in equation (7), the resolving time, τ , was found to be 87 μ sec.

RESOLUTION LOSS CURVE FOR THE

GAS COUNTER



B. Gamma radiation measurement

Electromagnetic radiations (gamma and X-rays) can be detected by means of the scintillations they produce when they pass through certain crystalline substances called 'phosphors'.

A photo-multiplier converts the scintillations into electrical pulses and these are recorded on appropriate scaling equipment after suitable amplification and selection. The electrical pulses are proportional to the intensity of the light that causes them, while the latter depends on the type of interaction by which the energy of the gamma rays is dissipated in the phosphor. The energy dissipation takes place in the three following ways:

i. Photo-electric effect:

The gamma ray ejects a bound electron from an atom or molecule and imparts to it a kinetic energy $(h \checkmark - b)$ where $h \checkmark$ is the energy of the photon and b the energy by which the electron is bound. By this process the photon is entirely absorbed, while the ionization produced through the interaction of this secondary electron results in a flash of light, the intensity of which is proportional to the amount of energy lost. If all the energy carried by this secondary electron is absorbed by the phosphor, then the light emitted is the maximum that can be produced for this particular gamma ray and a photopeak results.

- 83 -
ii. Compton scattering:

The photon, instead of giving up its entire energy to a bound electron, may transfer only part of its energy to an electron either bound or free. Thus the photon is not only degraded in energy but also deflected from its original path and may escape from the crystal either without any further loss of energy, or after it has undergone a number of scatterings, depositing part of its energy in each step. The light flashes produced by this process (Compton multiple scattering) will thus vary in intensity, causing a wide distribution of pulses in the photomultiplier.

It is also possible that the successive scatterings will stop the photon entirely within the phosphor, in which case the light emitted will again be seen by the photomultiplier as a photopeak.

iii. Pair production:

This process occurs only when the gamma rays have energies higher than 1.02 Mev, which is exactly the amount of energy required to raise an electron from a negative to a positive energy state, (1.02 Mev = $2m_oc^2$, where m_o is the rest mass of the electron) in which case an electron-positron pair is formed. The energy in excess of 1.02 Mev appears as kinetic energy shared by the two particles formed. When the positron is stopped, annihilation with a nearby electron occurs and two gamma quanta are released, each having an energy of 0.511 Mev. These annihilation gammas may or may not be detected by the phosphor. A characteristic 0.511 Mev photopeak in the pulse spectrum appears when they are fully absorbed by the phosphor.

A large variety of phosphors exists suitable for the detection of all types of radiation. For gamma radiation, the most efficient phosphor, and consequently the most widely used, is a NaI crystal activated by addition of the order of 0.1% TII. It is usually referred to as a NaI(Tl) crystal. This crystal was the detector in both types of instruments used in the present work for the measurement of the gamma radiation emitted by the nuclides studied.

(1) 100 Channel pulse height analyser

a. Equipment

The detector in this instrument consisted of a commercially available (Harshaw Chemical Co.) $3^{H} \times 3^{H}$ NaI(Tl) crystal hermetically sealed, and optically coupled to a Du-Mont type $6364,5^{H}$ photomultiplier tube. The high voltage was supplied to the photomultiplier from a Baird Atomic (model 318) stabilized high voltage supply unit. The photomultiplier tube was shielded from magnetic fields, which in general produce slight disturbances in its operation, by a mu-metal shield. The background radiation was reduced by placing the crystal-photomultiplier assembly into a lead cylinder $l_{\Xi}^{\pm H}$ thick. The fluorescent X-rays from the lead

were attenuated by lining the inside of the cylinder with a 'graded' shield, consisting of a $\frac{1}{2}$ iron lining and $\frac{1}{8}$ lucite. The detector assembly and the shielding, together with the geometrical arrangement used for the counting of the sources, are shown in Fig. 22. The electrical pulses from the photomultiplier were fed via a cathode follower (Hamner Electronics, model N-351) into a linear non-overloading amplifier (Baird Atomic, model 215) with variable gain. The positive output pulses were passed through an anode follower into a commercial 100 channel pulse height analyser (Computing Devices of Canada Limited, Model AEP 2230). The analyser was equipped for magnetic core storage of the data and oscilloscope presentation of the measured pulse height The stored data could be removed by external spectrum. equipment such as a pen recorder (Westronics Inc., Model 2705, TM 100-X84M-Y) accepting analog signals from the pulse height analyser and a digital print-out system which consisted of a print control unit (C.D.C. model 460), decimal scaler unit (C.D.C. model W 450), and the printer. Such an assemblage has a long resolution time, and the dead-time losses are serious when high counting rates are measured. The pulse height analyser was, however, equipped with a microammeter which indicated these dead-time losses. The dead-time losses were kept to a reasonable percentage by varying the geometrical efficiency under which the sources were measured. It should be mentioned here that these losses did not distort the spectrum but only reduced its

SCHEMATIC DIAGRAM OF DETECTOR ASSEMBLY INCLUDING DETECTOR, SOURCE GEOMETRICAL ARRANGEMENT, PHOTOMULTIPLIER, AND SHIELDING

.



overall amplitude. A block diagram of the pulse height analyser assembly is shown in Fig. 23.

b. Analysis of spectra

The data obtained from a pulse height analyser are given in the form of a pulse height spectrum.

In obtaining the gamma emission rate of a source from its pulse height distribution, all events which take place during the passage of the gamma radiation through the crystal must be taken into account. These include the photoelectric effect which appears as a characteristic photopeak, the Compton distribution, and the pair production if the gamma ray energy is above 1.02 Mev. This really means integration of the whole pulse height spectrum. In the case of multichannel instruments, the integration is achieved by adding the counting rates recorded in each individual channel. However, this process is not only tedious but is also subject to large errors even for simple spectra including no more than one characteristic photopeak, let alone the cases where more than one characteristic gamma is emitted by a nuclide or more than one nuclide is present in the source. The main difficulties arise from pulses due to radiation scattered by the radiation shield, beta absorbers or other material in the vicinity of the detector.

A simpler method consists in measuring the total number of events falling under the photopeak and converting those events into the absolute gamma emission rate. The conversion is done by the following relationship:

.....

BLOCK DIAGRAM OF PULSE HEIGHT

ANALYSER ASSEMBLY



$$N_{o} = \frac{N_{p}}{\boldsymbol{\ell} \times P \times A} \qquad (8)$$

- where No is the number of gamma rays emitted by a particular source.
 - N_p is the number of photoelectric events (area under the photopeak).
 - E is the total efficiency of the crystal defined as the probability of obtaining any size pulse when a gamma ray strikes the crystal.
 - P is the peak-to-total ratio which indicates what fraction of the total number of events will fall under the photopeak.
 - A is the correction factor for absorption in the source and any other absorbing material used in the measurement.

Of these factors, \mathcal{E} is determined theoretically and depends on the energy of the gamma radiation and the sourceto-detector geometrical arrangements. Heath (99) and Lazar⁽¹⁰⁰⁾ calculated the theoretical total efficiencies for a cylindrical 3" x 3" NaI(Tl) crystal for various source to crystal distances. P is determined experimentally from the product ($P \times \mathcal{E}$) which is called 'photopeak efficiency' and denotes the probability of obtaining a 'full energy' pulse if a gamma ray strikes the crystal. Both Heath and Lazar have reported peak to total ratios as a function of gamma ray P varies also with the size and shape of the energies. crystal, but source to detector geometry does not affect it. A, the absorption correction factor, is not a standard It depends on the type of sources used in any quantity. particular experiment and the absorbing material.

The sources used in this Laboratory for gamma radiation measurements were solutions of 2 ml volume enclosed in small standard screw cap vials. The measurement of the absorption factor for this type of source is quite a difficult task. In addition, constructional details in the crystal assembly rendered evident that, even if A were measured for all possible absorption effects, the reported efficiencies by Heath (99) and Lazar (100) could not be applied. An experimental determination of these three factors combined was therefore made in this Laboratory by M. May, G.V.S. Rayudu and G.R. Grant over an extended gamma ray energy range. The method has been described in detail by G.R. Grant⁽¹⁰¹⁾. The results were given in the form of combined photopeak efficiency curves for the various shelves of the geometrical assembly used.

In this work only Na^{24} and Cu^{64} were measured on the 100 channel pulse height analyser because it was not possible to obtain the C^{11} in a solution form convenient for later analysis of the carbon yield. Both Na^{24} and Cu^{64} sources were measured on shelf number zero. In estimating the area of the photopeak, the individual counts in each channel under it were added and the background subtracted. The background was estimated in the same way as it was done by the group that standardized the instrument⁽¹⁰¹⁾. Figs. 24 and 25 show typical spectra obtained for Cu^{64} and Na^{24} respectively. The dotted lines on these spectra indicate the estimated background in each individual case. Two

TYPICAL GAMMA RAY SPECTRUM FOR c_u^{64}



92a

TYPICAL	GAMMA	RAY	SPECT	RUM	FOR	Na ²⁴
(Inclu	uding	BOMe	Na ²²	acti	vit	r)



things, regarding the analysis of these spectra and the determination of the absolute gamma emission rates of the above nuclides, should be mentioned here.

i. Na²² is generally produced along with the Na²⁴ when The activity of aluminium is bombarded by protons. Na²² will be present in the measured sources. Since Na²² is a positron emitter⁽⁸⁹⁾, its 0.511 Mev gamma ray will obscure the pair production photopeak and, in addition, it will increase the 'coincidence summing' effect. Also, its 1.28 Mev gamma ray is much too close to the 1.368 Mev one of Na²⁴ and therefore impossible to be resolved from the latter. since the resolving ability of the detector for the 1.368 Mev photopeak was 16% (calculated from the spectra obtained in the course of this work). Hence the 1.368 Mev peak shown in Fig. 25 contains a contribution from the 1.28 Mev gamma ray of Na²². However, this contribution is rather small due to the fact that Na²² production is generally very small. Furthermore, Na²² has a long half-life (2.58 years) compared to that of Na²⁴ (14.9 hours) and thus the resulting counting rate for this nuclide is quite low. Hence its presence does not affect the shape of the 1.368 Mev photopeak, at least at the beginning of the measurement. Only towards the end of the decay of Na²⁴ was the shape of the photopeak distorted. Τo account for this, analysis of the 1.368 Mev photopeak

was carried out until it started showing this
distortion. Then the source was put aside until
Na²⁴ had decayed. The Na²² contribution was then
measured and subtracted from the gross decay curve.
ii. As already mentioned, all sources of Cu⁶⁴ and Na²⁴
were measured on the zero shelf of the detector
assembly. At such a high geometrical efficiency, the
'coincidence summing' effect is quite important^(99,100)
and should be accounted for in the analysis of the
spectra. However, because the 'summing' effect is
a rather complicated one to evaluate, the general
tendency in pulse height analyses is to measure the
gamma-ray sources in a sufficiently low geometrical

In most of the cases encountered in this work, time considerations did not permit measurement of the sources under these conditions, and therefore application of the standard combined efficiency curves determined in this Laboratory⁽¹⁰¹⁾ led to consistently low absolute gamma emission rates by 10%, since summing corrections were not made. The combined photopeak efficiencies in which the summing effect was not included were redetermined for the two particular nuclides studied. The average value for the 0.511 Mev photopeak of Cu⁶⁴, calculated from 15 different measurements, was found to be 0.0973 ± 0.0058 , while that for the 1.368 Mev photopeak of Na²⁴ was 0.0381 \pm 0.0022. These photopeak efficiency values were used in the present work.

(2) <u>Well-type scintillation counter</u>

a. Equipment

This was a commercial instrument built by Atomic Instruments Co. The detector part (model 810A) consisted of a NaI(T1) crystal 2" high and $1^{3}/4$ " in diameter with a centre well 5/8" in diameter and $1\frac{1}{2}$ " deep, optically coupled to a Du Mont, type 6292, photomultiplier tube. The photomultiplier pulses were amplified by a built-in amplifier and were then fed to the scaler (model 131) which had a built-in power supply unit (model 381). It was also equipped with a discriminator unit sensitive to negative pulses ranging from 0.25 - 10 volts, and a preset time clock. A stable high voltage supply (model 312) provided the positive high voltage to the photomultiplier. A block diagram of the instrument is shown in Fig. 26.

b. Calibration

Unlike the pulse height analyser, this counter cannot resolve pulses originating from different gamma ray energies or even distinguish between photoelectric events and Compton scattering. It simply records all events taking place in the detector crystal regardless of their origin. By varying the bias, it is possible to discriminate only against pulses that are smaller than a certain desired size. With a calibrated discriminator scale, a pulse distribution spectrum can be obtained in some simple cases. However, the best way to use this type of counter for quantitative work is to obtain an overall conversion factor for each individual

BLOCK DIAGRAM OF WELL-TYPE

SCINTILLATION COUNTER ASSEMBLY



nuclide to be studied, provided the source geometry remains within certain limits. This is actually the procedure adopted in the present work.

The sources of Na²⁴ and Cu⁶⁴ were those used with the pulse height analyser, i.e. solutions of 2 ml volume in standard glass vials. C^{11} was introduced into the same type of vials but in the form of BaCO₃. No geometry effect had to be used for Na²⁴ and Cu⁶⁴ sources since they were all of the same size. The geometry effect was checked in the case of C^{11} because small variations in the volume occupied by the BaCO₃ were expected. This effect was studied by placing a drop of a pure positron emitter (Ti⁴⁵ in this case) in a glass vial and determining the count rate of the source. A known amount of water was then added to the source in order to increase its volume, and the count rate was redetermined. The procedure was repeated until the initial source volume was brought up to 3 ml. No change in the count rate was detected up to a volume of 2 ml.

The overall conversion factors for Na^{24} and Cu^{64} were determined from data obtained in this work. The count rates of these sources, after suitable corrections for the presence of long-lived activity (Na^{22} in the case of Na^{24}) and dilution, were compared with the corresponding total disintegration rates obtained from 4π measurements. Since no 4π measurement data were available for C^{11} , the overall conversion factor in this instance was determined by comparison with the gas counting data. The overall conversion factors obtained are shown in Table V.

Nuclide	⁴ N /Scintillation Counter Conversio Factor	n Remarks
Na ²⁴	2.12 ± 0.148	Average of 13 measurements
Cu ⁶⁴	8.505 ± 0.648	Average of 11 measurements
oll	2.175 ± 0.119	Average of 14 measurements

Table V

It should be mentioned here that the decay schemes were not taken into account in the calculation of these factors.

The dead-time losses are also an important factor in the calibration of this counter. Dr. A. Kjelberg⁽¹⁰²⁾ had determined the dead-time losses and found that up to 300,000 c.p.m. no correction for these losses was necessary.

V TREATMENT OF DATA

Equation (4) (P. 2) clearly shows that the two key quantities necessary to determine the cross section of any nuclear reaction are the disintegration rate of the product nuclide and the beam intensity.

A. Disintegration rate determination

The disintegration rates at the end of bombardment of the nuclides studied were obtained by the following procedure:

1. The log of the measured counting rates of the sample

sources, corrected for resolution losses if necessary, were plotted versus time and the decay curves were thus constructed. Where long-lived activity was present (mainly in Na²⁴ and in some cases of Cu⁶⁴) the resulting gross decay curves were resolved in the usual graphical way into the single component decay curves which were then extrapolated back to the end of bombardment time. Typical decay curves for C¹¹, Cu⁶⁴ and Na²⁴, obtained in the course of the present work, are shown in Figs. 27, 28 and 29 respectively. The half-lives obtained for C¹¹ varied between 20.4 and 20.7 mins, for Cu⁶⁴ between 12.6 and 12.9 hrs and for Na²⁴ between 14.7 and 15.0 hrs.

An explanation is due here regarding the decay curves of C¹¹ obtained by gas phase measurements. Åв already mentioned, internal gas phase counting is sensitive to the presence of impurities. Van Slyke et al. (93) reported that the presence of impurities in the filling gas (vapours, air, etc.) slightly reduce the efficiency of the counter and also cause an increase in the plateau slope. Dr. R.A. Sharp⁽⁸⁰⁾ in his communication pointed out that either a vacuum of 10^{-4} to 10^{-5} mm Hg should be attained when the gas counter is evacuated or a waiting period would be necessary until these impurities are consumed and the counter characteristics become stable. The length of the waiting period would depend on the amount of

DECAY CURVE FOR THE 20.5 MINUTE C¹¹

•	- Gas Counter, measured activity
0	- Resolution loss corrections
	- Well-type Scintillation Counter



TIME AFTER BOMBARDMENT (MIN)

101a

DECAY CURVE FOR THE 12.8 HOUR Cu⁶⁴

□ - 4π Counter

△ - 100 Channel Pulse Height Analyser

O - Scintillation Counter



72

AFTER

48

TIME

96

BOMBARDMENT

120

144

(HRS)

10¹ 24

DECAY CURVE FOR THE 14.9 HOUR Na²⁴

ullet	-	Experimental points, 4π Counter
Δ	-	Resolved Na ²⁴ activity
0	-	Experimental points, Scintillation Counter (well type)
٨	-	Resolved Na ²⁴ activity
	-	Experimental points, Pulse Height Analyser
	-	Resolved Na ²⁴ activity



impurities present. A mass spectrometric analysis of the C.P. methane reported by Pate and Yaffe (90b) showed that some impurities were indeed present in the CH_{L} gas, ranging from $C_{2}H_{6}$ and higher unsaturated hydrocarbons to No and Oo which are the most injurious These impurities, although in very in this case. small percentages, were unavoidably introduced into the gas counter when the latter was filled with CHL and caused a small delay in the stabilization of the counter characteristics. This effect is clearly seen in the decay curve shown in Fig. 27, where a few of the initial counting rates, although corrected for resolution losses, still fall below the true half-life slope of the C¹¹ decay.

- 2. The counting rates at the end of bombardment were then converted to absolute counting rates by applying the correction factors associated with each method of measurement, as discussed in the appropriate sections of the Radiation Measurement Techniques. The total counting rate for each product nuclide was then obtained by multiplying these corrected counting rates by the appropriate dilution factors.
- 3. Finally, the disintegration rates were determined from the above data by taking into consideration the decay schemes of the nuclides studied, as applied to the particular method of counting used. Thus, in beta radiation measurements

where DR is the disintegration rate of the particular nuclide studied,

(CR)_β is the total absolute counting rate as determined by the beta radiation measurements, and

(BR)_B is the branching ratio for beta emission.

In the present work, the beta branching ratio for C^{11} and Na^{24} is $100\%^{(89)}$ while that for Cu^{64} is $57\%^{(89)}$ (19% β ⁺ and 38% β ⁻) since the electron capture branch was totally cut out. In gamma radiation measurements

$$DR = \frac{(CR)_r}{(BR)_r} (1 + \alpha_T) \dots (10)$$

where (CR), is the absolute emission rate for the particular gamma ray used,

(BR), is the fraction of the gamma transitions that this gamma represents (branching ratio), and

 α_{T} is the internal conversion coefficient of the same gamma ray.

The branching ratio for the 1.368 Mev gamma ray of Na²⁴ is 100% while that for the Cu⁶⁴ 0.511 Mev is 38% (i.e. 2 x 19%) since these gamma rays result from the annihilation of the positrons.

The internal conversion coefficients for both gamma rays were negligible and were not taken into account.

The counting rates obtained by the well-type scintillation counter were directly changed to disintegration rates by applying the overall conversion factors already mentioned. B. Beam intensity measurement

The beam intensity was monitored by means of the $C^{12}(p,pn)C^{11}$ reaction. A transformation of the general equation

$$(DR)_{p} = I n_{t} \sigma_{p} (1 - e^{-\lambda_{p} t}) \dots (Eq. 3, P. 2)$$

was worked out by applying it simultaneously for both target under study and monitor.

If subscripts (TS) and (PS) denote respectively target and product nuclides of the reaction under study and (TM) and (PM) the corresponding nuclides for the monitoring reaction, then

$$(DR)_{PS} = I n_{TS} \sigma_{PS} (1 - e^{-\lambda} PS^{t}) \dots (11)$$
$$(DR)_{PM} = I n_{TM} \sigma_{PM} (1 - e^{-\lambda} PM^{t}) \dots (12)$$

where (DR), I, n, σ , λ , and t have the same meaning given on pp. 1 & 2.

If equation (11) is divided by equation (12), the following expression is obtained:

$$\frac{(DR)_{PS}}{(DR)_{PM}} = \frac{n_{TS} \times \mathcal{O}_{PS} \times (1 - e^{-\lambda} PS^{t})}{n_{TM} \times \mathcal{O}_{PM} \times (1 - e^{-\lambda} PM^{t})} \dots (13)$$

where I is eliminated.

If one solves for the cross-section ratio G_{PS}/G_{PM} , equation (12) becomes

$$\frac{\mathcal{O}_{PS}}{\mathcal{O}_{PM}} = \frac{(DR)_{PS}}{(DR)_{PM}} \times \frac{n_{TM}}{n_{TS}} \times \frac{1 - e^{-\lambda}_{PM}t}{1 - e^{-\lambda}_{PS}t} \dots (14)$$

 n_{TM} and n_{TS} may be calculated from the experimental data in

the following way:

If W' is the total weight of any target foil of uniform thickness x, and A' is its total area, then the weight W of the target area exposed to the particle beam will be

$$W = W' \times \frac{A}{A'} = (SD) \times A \dots (15)$$

where (SD) is the superficial density of the target foil. The number, n, of nuclides contained in the exposed fraction of the target will then be

$$n = \frac{W}{(AW)} \times N \times (NA) = (SD) \times A \times N \times (NA) \dots (16)$$

where (AW) is the atomic weight of the target element used,

N is Avogadro's number, and

(NA) is the natural abundance of the particular isotope of the target element, which is of interest in the nuclear reaction under study.

If one substitutes n_{TS} and n_{TM} by their equivalents from equation (16) and takes into consideration that the area presented to the beam of incoming particles is the same for both target and monitor, equation (14) becomes

$$\frac{\tilde{O}_{PS}}{\tilde{O}_{PM}} = \frac{(DR)_{PS}}{(DR)_{PM}} \times \frac{(SD)_{TM}}{(SD)_{TS}} \times \frac{(NA)_{TM}}{(NA)_{TS}} \times \frac{(AW)_{TS}}{(AW)_{TM}} \times \frac{1 - e^{-\lambda} PM^{t}}{1 - e^{-\lambda} PS^{t}} \dots (17)$$

From equation (17) the cross-section ratios of the $Al^{27}(p,3pn)Na^{24}$ and $Cu^{65}(p,pn)Cu^{64}$ reactions to that of $C^{12}(p,pn)C^{11}$ were calculated. These ratios were subsequently converted into absolute cross-section values by using the results of Aamodt et al.⁽³⁶⁾ corrected according to Crandall

et al. (39) for the $C^{12}(p,pn)C^{11}$ excitation function. This excitation function, drawn on an expanded scale, is shown in Fig. 30.

C. Errors

Equation (17) may be written in a more convenient form $\vec{O}_{PS} = \vec{O}_{PM} \times K \times \frac{(DR)_{PS}}{(DR)_{PM}} \times \frac{(SD)_{TM}}{(SD)_{TS}} \times \frac{1 - e^{-\lambda}_{PM}t}{1 - e^{-\lambda}_{PS}t} \dots \dots \dots (18)$

where
$$K = \frac{(NA)_{TM}}{(NA)_{TS}} \times \frac{(AW)_{TS}}{(AW)_{TM}}$$
(19)

From equation (18) it is possible to calculate the standard deviation of the cross sections to be reported in this work. Let $\Delta(\mathbf{X})$ be the standard deviation of any of the measured quantities (X) involved in equation (18). Then

$$\left[\frac{\Delta \sigma_{PS}}{\sigma_{PS}}\right]^{2} = \left[\frac{\Delta \sigma_{PM}}{\sigma_{PM}}\right]^{2} + \left[\frac{\Delta_{K}}{\kappa}\right]^{2} + \left[\frac{\Delta(DR)_{PS}}{(DR)_{PS}}\right]^{2} + \left[\frac{\Delta(DR)_{PM}}{(DR)_{PM}}\right]^{2} + \left[\frac{\Delta(SD)_{TS}}{(SD)_{TM}}\right]^{2} + \left[\frac{\Delta(SD)_{TS}}{(SD)_{TS}}\right]^{2} + \left[\frac{\Delta(1 - e^{-\lambda}PM^{t})}{1 - e^{-\lambda}PM^{t}}\right]^{2} + \left[\frac{\Delta(1 - e^{-\lambda}PM^{t}}\right]^{2} + \left[\frac{\Delta$$

In the following paragraphs the standard deviation involved in each individual factor above will be discussed and quoted.

EXCITATION FUNCTION FOR THE

C¹²(p,pn)C¹¹ REACTION

(Expanded scale)

Results obtained from Aamodt et al. (36) and corrected as suggested by Crandall et al. (39)



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1. $\frac{\Delta \widetilde{O}_{PM}}{\widetilde{O}_{PM}}$

The above quantity indicates the error involved in the monitor cross-section values used. Aamodt et al. (36)quoted an error of $\pm 5\%$ in the absolute cross-section values reported. However, after applying the corrections suggested by Crandall et al. (39), it was not possible to accept that value any longer. An error of $\pm 10\%$ was adopted throughout the energy region of the McGill Synchrocyclotron. This is slightly pessimistic for the higher energies where the crosssection variation is slow, while it is optimistic for the low-energy region where the cross-section variation is rapid.

2. $\frac{\Delta_{K}}{K}$

No error was assumed for this term as it contains well-established values for the atomic weights of the elements used and their natural abundances, as obtained from the Nuclear Data sheets (89). If one wished to assign any error to that term, $\pm 1\%$ would be a rather pessimistic value since all three nuclides have been well studied.

3.
$$\frac{\Delta(DR)_{PS}}{(DR)_{PS}}$$
 and $\frac{\Delta(DR)_{PM}}{(DR)_{PM}}$

The standard deviations in the disintegration rates of the nuclides studied vary according to the methods of measurement used and the accuracy of their decay schemes. Thus, for the 4π measurement technique, which was the basic one in this work, the standard deviation in the disintegration
rate is determined from the following equation

$$DR = \frac{CR}{(BR) \times f_d \times f_s \times f_M \times f_{AN}} \qquad (21)$$

where CR is the count rate of the source,

- BR is the branching ratio of the nuclide studied,
- f_d is the dilution factor
- f, is the self-absorption correction factor,
- f_M is the source-mount absorption correction factor, and
- f_{AN} is the analytical factor which embraces the elution yield determination, by chemical analysis or other means.

The errors in the above factors can be kept small except for f_g and f_M . The error in these factors depended on their values. For a factor greater than 0.95, the error introduced is as low as 0.5%, while for lower values it increases. Of course the errors in the decay schemes cannot be avoided. For well-studied nuclides the errors are negligible. The errors adopted for the nuclides studied and the total estimated error in the corresponding disintegration rates are listed in Table VI.

Table VI

*		% erro:		%			
Nuclide	C.R.	B.R.	f _d	f s	f _M	fAN.	in D.R.
Na ²⁴	± 1	± 1	± 2		-	± 2	± 3
Cu ⁶⁴	± 1	± 2	<u>+</u> 2	<u>+</u> 2	<u>+</u> 2	± 3	± 5
c ^{ll}	± 1	± 1	-	± 3	-	± 3	± 4.2

The error in f for the case of C¹¹ was derived from the self-absorption study reported in Appendix I.

Similar calculations can be applied to all other methods of measurement in order to obtain the errors in the disintegration measurements. However, a simpler method for evaluating them was adopted, arising from the fact that the standardization of the equipment used was done against the 47 counter. By this standardization, all individual errors, such as aliquoting, analytical, decay scheme and efficiency, were combined in the overall conversion factors, determined by comparison of calculated absolute counting rates obtained in each case to the corresponding absolute disintegration rates determined by 4π measurements. Hence the statistical uncertainty in the disintegration rates obtained with all other methods will be the root-mean square of the errors in the 4π disintegration rates and the corresponding overall efficiency factors in each case.

These have already been mentioned in the appropriate sections of this work dealing with their determination. These errors are listed in Table VII.

	T	a	b	1	е	V	Ι	Ι	•
--	---	---	---	---	---	---	---	---	---

······································	%	Errors in th	he measuremen	t of
Nuclide	4រា DR	Gas Counter efficiency factor	Well-type Scin.Counter combined efficiency	Pulse Height Analyser combined efficiency
Na ²⁴	± 3	na nyanik Malakinin apali Kin apala pena pena Malakini dana p	± 7.0	± 5.8
Cu ⁶⁴	÷5	_	± 7.6	±6.6
011	±4.2	± 4.5	± 5.4	-

The overall uncertainties in the disintegration rate determinations are shown in Table VIII.

	% Errors	in disintegra	tion rate
Nuclide	Gas Counter	Well-type Scin.Counter	Pulse Height Analyser
Na ²⁴		± 7.6	± 6.5
Cu ⁶⁴	-	± 9.1	± 8.3
cll	± 6.1	± 8.2	-

Table VIII

4. $\frac{\Delta(\text{SD})_{\text{TM}}}{(\text{SD})_{\text{TM}}}$ and $\frac{\Delta(\text{SD})_{\text{TS}}}{(\text{SD})_{\text{TS}}}$

The errors for these quantities were experimentally determined. For Na and Cu foils the error was 1%, while that for C¹¹ was 3%, as already mentioned.

5.
$$\frac{\Delta(1 - e^{-\lambda_{\text{PM}}t})}{(1 - e^{-\lambda_{\text{PM}}t})} \text{ and } \frac{\Delta(1 - e^{-\lambda_{\text{PS}}t})}{(1 - e^{-\lambda_{\text{PS}}t})}$$

The errors involved in the determination of these values are difficult to calculate. They largely depend on the accuracy of the timing of the irradiations and the halflives of the product nuclides measured. For long-lived nuclides, small errors in the half-lives experimentally obtained do not affect the overall accuracy of the saturation terms. In the case of short-lived nuclides, large errors may result.

The intensity of the beam was assumed to be stable

during irradiation and the timing was done by means of a stopwatch. However, there is still an uncertainty regarding the stability of the beam intensity and an error of $\pm 2\%$ is assigned to the monitor saturation factor, while an error of $\pm 1\%$ was assigned to the saturation factors of Cu and Na, based on the good agreement obtained for the experimentally determined half-lives and their small decay constants.

On the basis of the above discussion and error assignments, the total error in the calculated cross sections of Na²⁴ and Cu⁶⁴ for every method of measurement was determined. Table IX lists the errors for each method of determination

Nuclide	Method of measurement	C ^{ll} Method of measurement	Total error (standard deviation) in calculation
N- 24	4 N	Gas Counter	± 13%
Na	Well-type Scin.Counter	Well-type Scin.Counter	± 16%
64	4T	Gas Counter	± 14%
Cu	Well-type Scin.Counter	Well-type Scin.Counter	± 17%

Table IX

An additional determination of the cross section of Cu^{64} , using Na²⁴ as a monitor, was determined for the pulse height analyser data obtained in this work. The monitor cross-section values used in this case were those determined

in the present work. The error in this determination was calculated and was found to be \pm 18%.

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RESULTS

The irradiations, performed in the McGill Synchrocyclotron, ranged between 21 Mev, which is just 2.5 Mev above the threshold of the monitor reaction, and 90 Mev, the maximum energy available.

In calculating the excitation functions of the $Al^{27}(p,3pn)Na^{24}$ and $Cu^{65}(p,pn)Cu^{64}$ reactions, the data were grouped in such a way that a direct check of the variations in the final result, caused by the use of different activity measurement techniques, would be possible. Since no pulse height analysis data were available for C^{11} measurement, the consistency of the results obtained by this method was checked by determining the excitation function of the $Cu^{65}(p,pn)Cu^{64}$ reaction relative to that of $Al^{27}(p,3pn)Na^{24}$. The monitor values used for this calculation were obtained from the excitation function of the above reaction, as determined by the other two methods of activity measurement.

Table X contains the disintegration rates of the product nuclides of each reaction, as determined by the various activity measurement techniques.

In Table XI the K values listed represent the combined result of all other experimental data which enter in the cross section calculations as required by Equation 17 (P. 107), that is

$$K = \frac{(AW)_{TS} \times (NA)_{TM} \times (SD)_{TM} \times (1 - e^{-\lambda} PM^{t})}{(AW)_{TM} \times (NA)_{TS} \times (SD)_{TS} \times (1 - e^{-\lambda} PS^{t})} \dots \dots (22)$$

The cross sections of the Al²⁷(p,3pn)Na²⁴ and Cu⁶⁵(p,3pn)Cu⁶⁴ reactions, calculated on the basis of C¹²(p,pn)C¹¹, are listed in Tables XII and XIII respectively. Each of these tables is followed by two graphical representations of the excitation functions obtained, one on linear and another on semilog scale. This was necessary, especially in the case of the Al²⁷(p,3pn)Na²⁴ reaction, in order to show clearly very low cross section values obtained near the threshold.

The cross-section values of the $Cu^{65}(p,pn)Cu^{64}$ reaction, calculated on the basis of the Al²⁷(p,3pn)Na²⁴ reaction from the pulse height analyser activity measurements, are shown in Table XIV. For comparison's sake, the same calculation was done using the 4π measurement data.

Fig. 35, following Table XIV, shows these results in comparison with the excitation function of the $Cu^{65}(p,pn)Cu^{64}$ reaction as obtained with respect to that of $Cl^{2}(p,pn)C^{11}$.

Whenever duplicate cross-section values were available, the average value was used in the construction of the excitation function. $E_p = Proton energy.$

G.C. = Gas counter.

W.S.C. = Well-type scintillation counter.

 $4\pi = 4\pi$ counter.

P.H.A. = 100 channel pulse height analyser.

a - No carbon foil was used in this bombardment.

b - Analysis of the carbon was spoiled.

- c The counter was not operating properly at the time of the measurement.
- d No Na²⁴ activity was produced at this energy.
- The Na²⁴ activity was so little that the spectra were not possible to analyse due to interference of Na²².
- f These irradiations were the first of the series and the Cu foil had not been included at the time.

		· · ·							
-	E	C.	11		Na ²⁴			Cu ⁶⁴	
No	r F . Mev	G.C.	W.S.C.	4π	W.S.C.	P.H.A.	4 A	W.S.C.	P.H.A.
1	21	1.693×10^9	1.711×10^9	đ	d	đ	4.527×10^8	4.733×10^8	4.300×10^8
2	27	7.780×10^9	7.659×10^9	1.550×10^4	1.604×10^4	e	4.507×10^8	4.457×10^8	4.307×10^8
3	33	6.589×10^9	6.164×10^9	1.800×10^5	1.537×10^5	2.052×10^5	1.279×10^8	1.138×10^8	1.325×10^8
4	33	1.342×10^{10}	c	5.894 x 10 ⁵	5.300 x 10 ⁵	5.638 x 10 ⁵	4.054×10^8	4.418×10^8	4.243×10^8
5	38	2.020×10^9	1.981 x 10 ⁹	2.070×10^5	1.855×10^5	1.929×10^5	4.165×10^{7}	3.768×10^7	3.835×10^7
6	43	8.295 x 10 ⁹	8.494 x 10 ⁹	3.450×10^6	3.286 x 10 ⁶	3.330×10^6	1.589×10^8	1.635×10^8	1.556×10^8
7	45.5	1.596×10^{10}	1.733×10^{10}	1.850×10^6	1.758×10^{6}	1.790×10^{6}	1.091×10^8	1.000×10^8	1.083×10^8
8	50	1.700×10^{10}	1.817×10^{10}	4.407×10^6	4.060×10^{6}	4.425×10^6	1.356×10^8	1.296×10^8	1.381×10^8
9	57 7	1.348 x 10 ⁹	Ъ	3.300×10^6	3.307×10^6	3.396 x 10 ⁶	f	f	ſ
10	62	1.956 x 10 ⁹	1.779×10^9	3.100×10^{6}	3.180 x 10 ⁶	2.959 x 10 ⁶	2.172×10^7	2.460×10^{7}	2.294×10^{7}
11	6 7 7	8.885×10^9	9.•375 x 10 ⁹	3.380 x 10 ⁷	3.328×10^7	3.529×10^7	f	f	f
12	72	1.307 x 10 ⁹	1.313 x 10 ⁹	3.730×10^6	3.604×10^6	3.398 x 10 ⁶	2.277×10^7	2.319×10^7	2.233×10^7
13	75	1.022×10^{10}	1.068×10^{10}	2.830×10^7	3.222×10^7	3.102×10^{7}	f	f	f
14	82	3.580 x 10 ⁹	Ъ	5.500×10^{6}	5.433 x 10 ⁶	5.326 x 10 ⁶	3.644×10^7	3.099×10^7	3.396×10^7
15	82	5.028x 10 ⁸	C	1.288×10^6	1.260×10^6	1.259×10^6	8.165x 10 ⁶	с	8.192x 10 ⁶
16	86.5	1.019 x 10 ⁹	1.058×10^9	3.100×10^6	3.180 x 10 ⁶	3.015×10^6	f	£	f
17/	86.5	8	8	1.790×10^{6}	1.759×10^{6}	1.744×10^{6}	1.072×10^{7}	1.118×10^{7}	1.043×10^{7}

<u>Table X</u>

DISINTEGRATION RATES OBTAINED BY THE VARIOUS METHODS OF ACTIVITY MEASUREMENT

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Table XI

CALCULATED CONSTANT FACTORS

v	=	(AW) _{TS}	~	(NA) _{TM}	~	(SD) _{TM}	•	1	-	e	$\lambda_{\rm PM} t$
	-	(AW) _{TM}	~	(NA) _{TS}	^	(SD) _{TS}	~	1	-	e	λ _{PS} t

Irradi-	Duration	K						
No.	Hours	Na^{24} vs. C^{11}	Cu^{64} vs. C^{11}	Cu^{64} vs. Na^{24}				
1	1.183	d	93.550	1.811				
2	0.980	58.887	106.570	1.809				
3	0.500	84.380	160.913	1.906				
4	0.750	65.056	124.685	1.916				
5	0.500	84.380	160.913	1.906				
6	0.500	89.625	174.205	1.943				
7	0.500	270.994	519.130	1.915				
8	0.500	167.410	320.689	1.915				
9	0.250	61.450	f	f				
10	0.500	89.625	177.598	1.981				
11	0.533	48.099	f	f				
12	0.758	67.814	118.340	1.745				
13	0.266	63.306	f	f				
14	0.392	119.680	215.943	1.874				
15	0.500	87.459	167.539	1.915				
16	0.308	61.176	f	f				
17	0.250	a.	a	1.740				

Irr	E		Disintegratio	n rate ratios	ocli	⁰ Na ²⁴	(mb)
No.	Mev	K	4π vs. GC	WSC vs. WSC	(mb)	4π vs. GC	WSC vs. WSC
1	21	đ	đ	a a	đ	đ	đ
2	27	58.887	1.992×10^{-6}	2.094×10^{-6}	58.5	0.007 ± 0.001	0.007 ± 0.001
3	3 3	84.380	2.732×10^{-5}	2.493×10^{-5}	80.0	0.184 ± 0.026	0.168 ± 0.028
4	33	65.056	4.390×10^{-5}	C	80.0	0.224 ± 0.031	C
5	38	84.380	1.025×10^{-4}	9.363 x 10 ⁻⁵	86.0	0.744 ± 0.104	0.680 ± 0.110
6	43	89.625	4.159×10^{-4}	3.868×10^{-4}	87.0	3.23 ± 0.45	3.00 ± 0.49
7	45.5	270.994	1.159×10^{-4}	1.015×10^{-4}	87.0	2.73 ± 0.38	2.40 ± 0.39
8	50	167.410	2.592×10^{-4}	2.234×10^{-4}	86.5	3.75 ± 0.52	3.20 ± 0.53
9	57	61.450	2.448×10^{-3}	ъ	82.5	12.4 ± 1.73	Ъ
10	62	89.625	1.585×10^{-3}	1.787×10^{-3}	79 •5	11.3 ± 1.6	12.7 ± 2.1
11	67	48.0 99	3.804×10^{-3}	3.550×10^{-3}	77.5	14.2 ± 2.0	13.2 ± 2.2
12	72	67.814	2.854×10^{-3}	2.745×10^{-3}	74.5	14.4 ± 2.0	13.8 ± 2.3
13	75	63.306	2.769×10^{-3}	3.017×10^{-3}	73.0	12.8 ± 1.8	13.8 ± 2.3
14	82	119.680	1.536×10^{-3}	Ъ	70.0	12.8 ± 1.8	Ъ
15	82	87.459	2.562×10^{-3}	c	70.0	15.6 ± 2.2	œ
16	86.5	61.176	3.042×10^{-3}	3.005×10^{-3}	68:0	12.6 ± 1.7	12.4 ± 2.0
17	86.5	a	· 8 ·	a (a ."	â	â

 $\frac{\text{Table XII}}{(p, 3pn) Na^{24}} \text{ REACTION USING } C^{12}(p, pn) C^{11} \text{ AS MONITOR}$

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4π versus G.C. data



Well-type scintillation counter data

Results of Yule and Turkevich (63)
Results of Hicks, Stevenson, and Nervik (59)

---- Excitation function of Hintz and Ramsey⁽³⁵⁾ corrected according to Crandall et al.⁽³⁹⁾



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SEMILOGARITHMIC PLOT OF THE EXCITATION FUNCTION OF THE A1 27 (p, 3pn) Na 24 REACTION OBTAINED ON THE BASIS OF THAT OF C¹²(p,pn)C¹¹ 4**M** versus G.C. data Well-type scintillation counter data Results of Hicks, Stevenson, and Nervik⁽⁵⁹⁾ Excitation function of Hintz and Ramsey⁽³⁵⁾ corrected according to Crandall et al. (39) Yule and Turkevich⁽⁶³⁾



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CROS	SS SECT	ION CALCULA	TIONS FOR THE C	⁰ ^{(p,pn)Cu⁰⁴}	REACTION USING	$C^{\perp 2}(p, pn)C^{\perp}$	AS MONITOR
			Disintegratio	on rate ratios	σ_11	o _{Cu} 64	(mb)
Irr No.	^г р Mev	K	4π vs. GC	WSC vs. WSC	(mb)	4π vs. GC	WSC vs. WSC
1	21	93.550	2.674×10^{-1}	2.765×10^{-1}	13.5	338 [±] 51	349 [±] 61
2	27	106.570	5.793×10^{-2}	5.819 x 10^{-2}	58.5	361 ± 54	363 ± 64
3	33	160.913	1.941×10^{-2}	1.846×10^{-2}	80.0	249 ± 37	238 ± 42
4	33	124.685	3.020×10^{-2}	c	80.0	301 ± 45	Č
5	38	160.913	2.062×10^{-2}	1.902×10^{-2}	86.0	285 ± 43	263 ± 46
6	43	174.205	1.915×10^{-2}	1.924×10^{-2}	87.0	290 ± 43	2 <u>9</u> 1 ± 51
7	45.5	519.130	6.830×10^{-3}	5.769×10^{-2}	87.0	308 ± 46	260 ± 46
8	50	320.689	7.976×10^{-3}	7.133×10^{-2}	86.5	221 ± 33	198 ± 35
10	62	177.598	1.110×10^{-2}	1.382×10^{-2}	79•5	157 ± 23	195 ± 34
12	72	118.340	1.742×10^{-2}	1.764×10^{-2}	74.5	154 [±] 23	156 ± 27
14	82	215.943	1.018×10^{-2}	Ъ	70.0	154 ± 23	Б
15	82	167.539	1.623×10^{-2}	С	70.0	190 ± 28	C

					Table	XIII					
ROSS	SECTION	CALCULATIONS	FOR	THE	$Gu^{65}(n,m)$)Cu 64	REACTION	USING	$C^{12}(p,pn)C$	11 AS	MONITO

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LINEAR PLOT OF THE EXCITATION FUNCTION OF THE $Cu^{65}(p,pn)Cu^{64}$ REACTION, OBTAINED BY USING REACTION $\frac{C^{12}(p,pn)C^{11}}{C^{12}}$ AS MONITOR

4 N vs. G.C. data

Well-type scintillation counter data

Excitation function of Meadows⁽⁶⁵⁾, corrected
Results of Yule and Turkevich⁽⁶³⁾
Results of Coleman and Tewes⁽⁶⁷⁾



SEMILOGARITHMIC PLOT OF THE EXCITATION FUNCTION OF THE Cu⁶⁵(p,pn)Cu⁶⁴ <u>REACTION OBTAINED BY USING</u> <u>c¹²(p,pn)C¹¹ AS A MONITOR</u>



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▲

Well-type scintillation counter data

Excitation function of Meadows (65) corrected

Yule and Turkevich⁽⁶³⁾



				Table XIV			
		CROSS S	SECTION CALCULAT	IONS FOR THE Cu	⁶⁵ (p,pn)Cu ⁶⁴	REACTION	
			USING A12	⁷ (p,3pn)Na ²⁴ AS	MONITOR		
Trr	En	• <u>18-2</u> • • • • • • • • •	Disintegration	n rate ratios	a _{Na} 24	o _{Cu} 64	(mb)
No.	Mev	K	41 vs. 41	PHA vs. PHA	(mb)	4π vs. 4π	PHA vs. PHA
2	27	1.809	2.907 x 10 ⁴	8	0.007	368 + 59	θ
3	33	1.906	7.105×10^2	6.457×10^2	0.200	271 [±] 43	246 ± 44
4	33	1.916	6.878×10^2	7%525 x 10 ²	0.200	264 [±] 42	288 ± 52
5	38	1.906	2.012 x 10^2	1.988×10^2	0.700	268 ± 42	265 ± 48
6	43	1.943	46.06	46.72	1.600	143 ± 23	145 ± 26
7	45.5	1.915	58.97	60.50	2.300	260 ± 41	266 ± 48
8	50	1.915	30.76	31.21	4.300	247 ± 39	251 + 45
10	62	1.981	7.006	7.752	13.6	189 ± 30	209 ± 37
12	72	1.745	6.105	6.571	14.0	149 [±] 24	161 [±] 29
14	82	1.874	6.625	6.376	13.9)	173 ± 28	166 ± 30
15	82	1.915	6.339	6.506	13.9	169 [±] 28	173 ± 31
17	86.5	1.740	5.989	5.981	13.8	144 ± 23	144 ± 265

<u>GRAPHICAL REPRESENTATION OF THE CROSS</u> <u>SECTION VALUES FOR THE REACTION</u> <u>Cu⁶⁵(p,pn)Cu⁶⁴, OBTAINED ON THE BASIS OF</u> <u>THE A1²⁷(p,3pn)Na²⁴ REACTION</u> <u>EXCITATION FUNCTION SHOWN IN FIG. 31</u>

> Excitation function obtained on the basis of the C¹²(p,pn)C¹¹ reaction (Fig. 33)

O 47 counting data

Pulse Height Analyser counting data



DISCUSSION

I GENERAL

The discrepancies in the excitation functions of the reactions studied may be due to three sources of errors.

1. Errors originating in the irradiation techniques used

These can be of two kinds:

- (a) Positive errors due to artificial increase in the measured residual activity, from secondary reactions leading to the same reaction product.
- (b) Negative errors arising from losses in the residual activity due to nuclear recoil processes.

(a) Errors of this type exist in some of the earlier works using thick targets or the 'stacked foil' technique. The latter, described in detail by Aamodt et al. (36) and by Hintz and Ramsey (35), was widely used since it could yield a full excitation function in a single irradiation. The secondary particle production inherent in this technique was corrected for in most cases by applying a small correction factor to the experimental results. That large errors could be introduced by the above factor was first pointed out by Crandall et al. (39) who received further support from Rosenfeld et al. (42). The latter pointed out that secondary protons and neutrons with wide energy distribution are produced in the absorbers by inelastic collisions.

The secondary protons may either be stopped within the absorber in which they were produced or emerge from it with energies falling below or above the threshold of the nuclear reaction studied. The energy distribution of the emerging protons, and thus their contribution to the product activity, depends on the initial energy of the proton beam, the thickness of the absorbers (energy degradation steps) and the characteristics of the very same excitation function under study. Generally, this contribution is small and does not extend too far into the target stack, at least when the energy of the initial proton beam is of the order of 100 Mev⁽⁴²⁾.

The case is different with the secondary neutrons. In general, neutron-induced reactions have lower thresholds, higher cross sections, and their excitation functions have maxima occurring at rather low energies. This means that neutrons are more effective at low energies. As the proton beam penetrates into the stack, high-energy neutrons which did not cause secondary reactions are also degraded and add on to the new low-energy neutrons produced. Thus the errors due to secondary reactions induced by neutrons increase in a cumulative manner as the low-energy part of the target stack is approached, until the emerging neutrons reach the energy level of their maximum effectiveness. Beyond that level their effectiveness falls off rapidly. The net effect of this process is an apparent lowering in the threshold energy and a consequent raising of the lower

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portion of the excitation function.

The magnitude of the error introduced by these secondary reactions cannot be estimated unless flux and energy distribution of the secondary particles are known at every point in the stack.

The above discussion applies also when 'thick' targets are used, although the errors in this case are generally small. For instance, Crandall et al. (39) estimated that the secondary reaction contributions in the case of the Al²⁷(p,3pn)Na²⁴ reaction are less than 1% if the target thickness does not exceed 0.5 g/cm².

(b) Thin targets are in wide use today, and the 'stacked foil' technique has virtually been abandoned. However, the thin target technique may introduce some errors due to activity losses from nuclear recoil processes. The latter are due to momentum transfer and occur when nuclear collisions or emission of nucleons take place. The recoil losses depend on the range-energy relationship of the recoiling nuclei in the target material.

Fung and Perlman⁽¹⁰³⁾, for instance, bombarded thin Al foils (0.25 and 0.5 mil) with protons, deuterons, and alpha particles of various energies, and measured the amount of Na²⁴ activity lost in every case. They found that losses in the direction of the beam, presumably due to collisions of the target nuclei with the bombarding particles, were greater than the losses in the opposite direction, which were consistently below 2%. They also found that the losses in

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the forward direction increased as the masses and the energy of the bombarding particles increased. This increase was not monotonic but had a maximum which occurred at about 60-70 Mev in the case of protons, 120-140 Mev in that of deuterons and at about 160 Mev for the alpha particles. The maximum recoil loss for the 0.25 mil foil was 29%, while that for the 0.50 mil was 14.5%. These results indicate that the optimum thickness of a target to be used depends upon the type and energy of the bombarding particles. The errors due to secondary reactions define the upper limit of thickness to be used. An alternative method of eliminating recoil losses is to use a target consisting of three thin foils whereby losses occurring in the second foil are compensated by the contributions from the first and third.

2. Errors in the activity measurement techniques

These are inherent in the methods used and depend on the decay characteristics of the activity measured, the instruments employed, and the calibration factors associated with them. This discussion will briefly outline the possible errors in each of the methods of activity measurement used.

(a) External beta measurement techniques:

Most of the early work was done by techniques involving the use of Geiger-Müller or End-Window proportional counters. These had the advantage of often eliminating chemical treatment of the targets. Chemical treatment was done only when a large number of radioactive products resulted from the target bombardment (e.g. Cu bombardments). However, the results obtained by these measurement techniques were not so accurate because of the large number of correction factors needed to convert the measured counting rates to absolute rates. These factors are:

i. Geometrical efficiency of the counter.

ii. Absorption of activity in the air space between source and counter window.

iii. Absorption of activity in the counter window.

iv. Absorption and scattering of the radiation emitted by the source itself (self-absorption and

self-scattering).

v. Backscattering or radiation by the source support.

The last two factors are the most important of all and have been the objects of intensive studies⁽¹⁰⁴⁻¹⁰⁹⁾. Many disagreements in experimental results obtained by external beta measurement techniques were due to these two factors.

The cause of the disagreements was resolved when Seliger^(40,41) reported that the saturation backscattering, although independent of the energy of the beta radiation, was lower for positrons than for negatrons. This suggested that the self-absorption factor also should depend on the sign of the beta radiation besides its energy. The latter was verified experimentally by Rosenfeld et al.⁽⁴²⁾ in the case of C^{11} .

An estimated error of \pm (15-20)% has been repeatedly

attached to results obtained by these methods by various investigators. However, it must be borne in mind that the accuracy can be improved by employing better-quality instruments and more refined calibration procedures.

(b) Internal beta measurement techniques:

These techniques are an improvement in the activity measurements because the introduction of the active source inside the counter chamber removes all external calibration factors.

Three main types of countersare in wide use today, namely 2π , 4π , and gas phase. The correction factors necessary for the 2π counter are those of backscattering and self-absorption, while in the case of the 4π counter only the self-absorption factor is of importance. In regard to the gas counter, which can be used only when the source to be measured can be brought into the gas phase, even the self-absorption factor is eliminated.

Gas phase counting is particularly suitable for the measurement of soft beta radiation, for which the selfabsorption losses are quite high. Certainly, in this particular technique, other problems arise, as discussed elsewhere in this work. However, an accuracy within \pm 5% or better can easily be achieved. The accuracy of the 2 \Re counter is of the same order.

The 4π measurement technique has been the object of intensive studies (90,91,110-114) and the continuous refinements in the calibration procedures have rendered it one of the most accurate methods for activity measurements. Accuracy within ± 1% or better can easily be achieved if instruments of good quality are used and proper care in the calibration procedure is exercised.

(c) Gamma measurement techniques:

The errors in these techniques are different in nature. They are mainly due to the quality of the instruments used, the efficiency of the detection devices, the complexity of the gamma-decay scheme of the sources measured and the analysis of the spectra. The latter especially has been studied repeatedly, and various methods of analysis have been proposed whereby simplifications and improvements have rendered possible the measurement of gamma activity with an average accuracy of \pm (5-10)%. There is no upper or lower limit of error in this type of measurement. In cases of complex gamma spectra, the error increases, while for simple spectra the error may be even less than ± 5%. An improvement in the accuracy of this technique is possible by calibrating the instrument with sources previously standardized on a $4 \Re$ counter.

3. Errors originating from the beam monitoring

No limit can be set for these errors. They depend entirely on the method by which the beam is monitored. If it is done by absolute means, the error depends on the accuracy inherent in the instrumental arrangement employed.

In cases where relative measurements are utilized, proper selection of the monitor reaction is a very important

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factor. The accuracy of the monitor cross-section values affect to a great extent that of the reaction studied. One should also bear in mind that in any nuclear reaction the cross sections change rapidly in the energy range near the threshold. They are therefore less accurate for use as reference standards in this energy range.

II CONSISTENCY OF RESULTS REPORTED IN THIS WORK

From the preceding discussion, it is obvious that results obtained, even by the same experimenter, may vary considerably if different experimental techniques are employed. One of the aims of this work was to check the magnitude of these variations arising from the use of different activity measurement techniques. Any errors in the activity measurements are reflected in the final calculation of the disintegration rate ratios and therefore in the determined cross sections. Tables XII, XIII and XIV (pp. 121, 124 and 127 respectively) provide a direct measure of these variations.

An 'average' deviation between the results obtained by the different activity measurement techniques in this work was calculated in the following manner:

The disintegration rate ratios obtained by beta measurements were arbitrarily considered as standard. From these the corresponding ratios, obtained by gamma measurements, were subtracted, the differences expressed as per cent deviation from the standard, and their root mean square extracted.

The 'average' deviation of the well-scintillation counting data was $\pm 8.9\%$ for the Al²⁷(p,3pn)Na²⁴ reaction excitation function (Table XII) and $\pm 10.8\%$ for that of the Cu⁶⁵(p,pn)Cu⁶⁴ reaction (Table XIII), while the 'average' deviation of the pulse height analyser data (Table XIV) was 5.8%.

Intercomparison of the different measurement techniques, as seen in Tables VI and VIII (pp. 111 and 113) shows that the 4π and gas counting techniques are the most accurate. The 100 channel pulse height analyser technique is less accurate than the 4π , but more accurate than that of the well-type scintillation counter. This explains the higher deviation obtained when comparing the 4π and gas counter data to those of the well-type scintillation counter, than when comparing the 4π data to those of the pulse height analyser.

III COMPARISON WITH OTHER LITERATURE RESULTS

1. Al²⁷(p,3pn)Na²⁴

The works of Hintz and Ramsey⁽³⁵⁾ and of Hicks, Stevenson, and Nervik⁽⁵⁹⁾ are the only ones that cover the energy region of interest. In both papers, as well as in this work, the $C^{12}(p,pn)C^{11}$ reaction was used as a monitor.

Hintz and Ramsey irradiated their targets by the 'stacked foil'technique. They also measured the resulting Na²⁴ and C¹¹ activities on a Geiger counter. As monitor

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values they used the results of Aamodt et al. (36), who had determined the excitation function of the $C^{12}(p,pn)C^{11}$ reaction, by employing the same irradiation and activity measurement techniques with the exception that they monitored their proton beam by absolute means. Crandall et al. (39) and Rosenfeld et al. (42) later proved that these results were high by 13%. The error was attributed partly to the irradiation and partly to the activity measurement techniques. Aamodt et al. ignored in the calibration of their counter the difference in the backscattering and self-absorption of positrons and negatrons (41, 42). Hintz and Ramsey calibrated their counter by obtaining a composite efficiency curve for each of the nuclides measured. Thus they avoided the error that entered into the results of Aamodt et al. Their results then would be high only by 13%, due to the monitor values employed, if no errors from the irradiation technique were introduced in the production of Na²⁴.

In Fig. 31 are shown the corrected results of Hintz and Ramsey, those of Hicks, Stevenson, and Nervik, and the excitation function obtained in this work. Two additional cross-section values from the work of Yule and Turkevich⁽⁶³⁾, which fall within the energy region studied, are also included. As it can be seen, a very close agreement exists between the results of Hintz and Ramsey and those obtained in this work above 55 Mev. Below this energy, however, the results of Hintz and Ramsey are higher, and there is also disagreement in the apparent threshold of the reaction. Both the results of Hicks, Stevenson, and Nervik, and those of Yule and Turkevich are considerably lower throughout the energy region studied.

The results reported in this work are believed to be free of any systematic errors due to the irradiation technique used by Hintz and Ramsey. Therefore the good agreement with their high-energy results indicates that between 55 and 100 Mev the errors due to secondary protons and neutrons inherent in the stacked foil technique are not appreciable for this reaction. This is in agreement with the predictions of Rosenfeld et al. (42)

The disagreement in the low-energy results and the threshold energy can be attributed to the $Al^{27}(n,\alpha)Na^{24}$ reaction induced by secondary neutrons in the manner described on P. 130. This reaction has a calculated threshold of about 3 Mev (on a Q basis alone).

An alternative way in which Na²⁴ may be produced is by the $A1^{27}(p,pHe^3)Na^{24}$ reaction having a calculated threshold of about 23 Mev. It is certainly present throughout the energy region studied. However, it is unlikely that this reaction may also be responsible to any extent for the disagreement below 55 Mev, since secondary protons do not reach that far in the stack⁽⁴²⁾.

A careful examination of the experimental methods used by Hicks, Stevenson, and Nervik⁽⁵⁹⁾ and by Yule and Turkevich⁽⁶³⁾ showed no apparent reason for the large difference between their results and those reported here. Errors in the irradiation techniques are ruled out as they both used the thin compensated target method.

No C¹¹ activity losses occurred in this work either. The carbon sources were thick enough to ensure against recoil losses and the chemical treatment was done on the target as a whole, thus avoiding any handling that could cause losses of a mechanical nature due to the brittleness of the carbon foils. The only potential source of difference may be found in the C¹¹ activity measurements. It is not likely that the C¹¹ activity measured in this work could be low because of the back extrapolation for 10 to 12 It is true that a 5% error in the half-life half-lives. slope (the difference between 20.5 min and 21.5 min) could result in an error of -30%. However, the good agreement in the half-lives (20.5 \pm 0.2), as well as in the final disintegration rates estimated by the two measurement techniques employed, rules out this possibility. On the other hand, an error may have been introduced in the intercalibration of the two end-window counters used by Hicks, Stevenson, and If they ignored the difference in the counting Nervik. efficiency of positrons and negatrons for this type of counter, the resulting disintegration rates of C¹¹ would be higher. This criticism does not apply to the work of Yule and Turkevich, and the small difference between their results and those of Hicks, Stevenson, and Nervik shows that this error cannot be too large.

A possible reason for the low values in both of

those works could be an artificial increase in the C^{11} activity due to the carbon source material used. Commercial polymers may contain various amounts of fillers and remnants of the catalysts used during the polymerization process. If any of these admixtures produced some activity having a half-life comparable to that of C^{11} , its contribution would be added to the measured disintegration rates of C^{11} , since the C^{11} sources did not undergo any chemical treatment. An activation analysis of their carbon source material could throw some light on this possibility.

The compatibility of the results obtained in this work with those of other investigators in the higher energy region was also checked. Fig. 36 shows results obtained by other investigators up to 1 Bev, including those of Hicks, Stevenson, and Nervik and of Yule and Turkevich. Some of them were determined by employing absolute beam monitoring techniques (39,57,64). The results of this work can be joined to those obtained in the high-energy region by a smooth monotonically decreasing line, a trend that is in keeping with the Serber mechanism which presumably predominates in the energy region above 100 Mev.

The excitation functions of Hicks, Stevenson, and Nervik and of Yule and Turkevich, as shown in this figure, in addition to being low go through a valley in the energy region between 140-260 Mev, whereupon their results become comparable to those of other investigators.

The cause of this valley has not yet been explained.

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<u>COLLECTED RESULTS FOR THE REACTION</u> A1²⁷(p,3pn)Na²⁴ FROM THRESHOLD TO 1 BEV

Excitation function reported in this work
 Proposed continuation for the high-energy region
 Crandall et al.⁽³⁹⁾
 Prokoshkin and Tiapkin⁽⁴⁷⁾
 Marquez⁽⁵⁷⁾
 Hicks, Stevenson and Nervik⁽⁵⁹⁾
 Yule and Turkevich⁽⁶³⁾
 Goebel and Shultes⁽⁶⁴⁾
 Friedlander et al.⁽⁶¹⁾



Unfortunately the data available in this energy region could not definitely confirm its existence or disprove it. One cross-section value reported by Prokoshkin and Tiapkin⁽⁴⁷⁾ at 150 Mev is definitely higher and falls on the proposed line joining the data of this work to those of the higher energy. If the existence of this valley is accepted, then a possible explanation could be given by assuming that, in the low-energy region, the major part of the Na²⁴ activity comes from the Al²⁷(p,pHe³)Na²⁴ reaction than from the Al²⁷(p,3pn)Na²⁴ reaction. As the bombarding energy increases the contribution of the former reaction decreases while that of the latter increases, resulting in this valley. However, this possibility has been ruled out by Hicks, Stevenson, and Nervik.

2. Cu⁶⁵(p,pn)Cu⁶⁴

Of the work reported in the literature, only the excitation function of Meadows⁽⁶⁵⁾ covers fully the energy region of interest. All other work has been concentrated in the high-energy region and only occasionally has a cross-section value below 100 Mev been reported^(63,67). Meadows measured the excitation function of this reaction relative to the $A1^{27}(p,3pn)Na^{24}$ reaction, using the uncorrected values of Hintz and Ramsey to calculate the absolute cross sections. He also used the 'stacked foil' technique and measured the resulting activities of Na²⁴ and Cu⁶⁴ on a Geiger counter. He applied the usual corrections including that for self-absorption and self-scattering. He ignored the backscattering

correction entirely, using the argument that any error due to the difference in the backscattering coefficient for positrons and negatrons was included in the results of Hintz and Ramsey. Since Cu^{64} is also partly a positron emitter, this error would be compensated for. This statement is not valid because the positron decay branch of Cu^{64} is only 19%. Crandall et al.⁽³⁹⁾ pointed out that a reduction of 13% should be applied to the results of Meadows. The corrected excitation function of Meadows, together with that obtained in this work, and some of the results reported by Coleman and Tewes⁽⁶⁷⁾ and by Yule and Turkevich⁽³⁾ are shown in Fig. 33.

Although with this correction the results of Meadows fall within the experimental error of this work, they still remain consistently higher. If one looks at the two excitation functions, it will be noticed that they run parallel above 40 Mev. An increasing deviation occurs in the energy region between 20 and 40 Mev. This work has not been extended below 21 Mev because the threshold of the monitor reaction did not permit it. An attempt at 18 Mev failed to produce any appreciable activity in the carbon target. Besides, the excitation functions of both the $Cu^{65}(p,pn)Cu^{64}$ and the $C^{12}(p,pn)C^{11}$ reactions fall off so rapidly below this energy that any result obtained would bear a very large error. The continuation of this excitation function below 21 Mev (dotted line) was based on the calculated threshold for the reaction under discussion, which is about 10 Mev and not 5 Mev as deduced from the work of Meadows. Furthermore, the cross-section values below 24 Mev, as given by him, are questionable. The threshold of the monitor reaction reported by Hintz and Ramsey is 24 Mev. Therefore the beam intensity below this energy could not have been estimated unless some other method had been used. It has not been possible to draw any conclusions on this score from the report of Meadows.

The large deviations of the low-energy results are due to the 'stacked foil' technique whereby secondary neutroninduced reactions produce additional Cu⁶⁴ activity in the manner already described on P. 130. Besides the $Cu^{65}(n,2n)Cu^{64}$ reaction, which has a threshold of 10 Mev, Cu^{64} may be produced via the $Cu^{65}(p,d)Cu^{64}$ reaction, which has a threshold of 7.5 Mev. However, the contribution of this reaction due to secondary protons is unlikely for reasons already discussed (P. 130). Even though an appreciable contribution to this reaction due to the primary proton beam has been ruled out by both Yule and Turkevich and Meadows, the low threshold reported by the latter may well be attributed to this reaction. In any case, it cannot be held responsible for the differences in the results, even in the higher energies. It is more probable that secondary neutrons may still be partly responsible for this deviation since the cross section of the $Cu^{65}(n,2n)Cu^{64}$ reaction at 90 Mev is 98 mb⁽⁶⁷⁾, that is comparable to that of the (p,pn) type.

The manner in which secondary reactions affect the

results may be clearly seen in Fig. 37. Curve (a) represents the differences in the cross-section values of Hintz and Ramsey and of this work for the $Al^{27}(p, 3pn)Na^{24}$ reaction. Curve (b) is the same type of curve obtained by comparing the Cu⁶⁵(p,pn)Cu⁶⁴ reaction excitation functions of Meadows and of this work.

The low cross-section values reported by Yule and Turkevich for the 100-Mev region can be explained on the same terms as their results for the Al²⁷(p,3pn)Na²⁴ reaction. The cross-section value at 90 Mev of Coleman and Tewes⁽⁶⁷⁾ agrees within experimental error with that of this work. Furthermore, as it can be easily seen in Fig. 38 where data from the work done in the high-energy region are also shown, the excitation function reported here can be joined to them quite well, if some of the low-energy results of Yule and Turkevich are neglected.

GRAPHICAL REPRESENTATION OF SECONDARY REACTION CONTRIBUTIONS IN THE STACKED FOIL TECHNIQUE

- (a) Contribution from the $Al^{27}(n, \propto) Na^{24}$ reaction to that of $Al^{27}(p, 3pn) Na^{24}$
- (b) Contribution from the $Cu^{65}(n,2n)Cu^{64}$ reaction to that of $Cu^{65}(p,pn)Cu^{64}$





COLLECTED RESULTS FOR THE Cu⁶⁵(p,pn)Cu⁶⁴ REACTION FROM THRESHOLD TO 1 BEV

- ----- Excitation function reported in this work
- - Proposed continuations for high and low energy regions
 - ♦ Batzel et al. (10)
 - ▲ Yule and Turkevich (63)
 - Markowitz et al. (70)
 - Coleman and Tewes
 - Vinogradov
 (71)



SUMMARY

The excitation functions of the reactions Al²⁷(p,3pn)Na²⁴ and Cu⁶⁵(p,pn)Cu⁶⁴, often used as monitors, have been determined relative to that of $C^{12}(p,pn)C^{11}$ from 20 - 90 Mev.

Irradiations were performed in the internal beam of the McGill Synchrocyclotron.

Target thicknesses were chosen so as to minimize secondary particle production and recoil losses.

The resulting activities of C^{11} , Na^{24} and Cu^{64} were measured by employing various techniques in order to determine their effect on the cross-section measurements. The deviations observed ranged from \pm 6% to \pm 11%, depending on the accuracy of the techniques used. This indicates that discrepancies of this order may be expected in reported results even if all other types of experimental errors have been rendered negligible.

In the case of the Al²⁷(p,3pn)Na²⁴ reaction, the results reported in this work were found to be in close agreement with those of Hintz and Ramsey above 55 Mev when the latter were reduced by 13% to account for the monitor values they employed. Below 55 Mev, the results of Hintz and Ramsey were high. In contrast, those of Hicks, Stevenson, and Nervik were found to be too low, but no definite explanation for this discrepancy was found. With regard to the $Cu^{65}(p,pn)Cu^{64}$ reaction, the results obtained here were found to be consistently lower than those of Meadows, even when the latter were reduced by 13% for the same reasons as in the case of Hintz and Ramsey.

The observed differences, although within experimental error in the region above 40 Mev, increased sharply between 20 and 40 Mev. These low-energy discrepancies in both cases were explained in terms of neutron-induced secondary reactions, owing to the irradiation techniques used by Hintz and Ramsey as well as by Meadows.

Experimental

In determining the efficiency of the gas-counter system employed in this work, it became necessary to determine the absolute disintegration rates of C^{11} activity by the 4 π beta measurement technique (pp. 78 & 79).

This section is concerned with the standardization of the 4π counter so that good accuracy could be achieved in these measurements. The factors involved in the standardization of this counter have been described in detail by Pate and Yaffe^(90b-90e). In the case of C¹¹ the selfabsorption correction was the most important since the sources used were thick. These sources were prepared by bombarding carbon foils in the McGill Synchrocyclotron, which were then burnt by the method described on P. 49.

The resulting CO₂ was converted to BaCO₃ and sources were prepared by filtering various amounts of BaCO₃ by means of the suction apparatus shown in Fig. 39. The sources, all of approximately equal diameter and of good uniformity, were carefully washed and dried on the filter paper which was placed on the fritted disc. When dry, the sources did not adhere to the filter paper and therefore it was easy to transfer them on to VYNS films by simply placing the film ring on top of the source, turning it upside down and then removing the filter paper. The source was then covered by another VYNS film to secure it against moving.

SUCTION APPARATUS USED FOR THE

PREPARATION OF THE BaCO3 SOURCES



The thickness of these films never exceeded $15 \ \mu g/cm^2$, thus rendering source-mount absorption corrections negligible. The count rates of the sources were measured and corrected for decay to some common arbitrarily chosen zero time. The source area was measured, and the mass was determined by the volumetric method described on P. 55.

The specific activities of these sources were plotted against their respective superficial densities and the resulting curve was extrapolated to zero. Then the measured specific activities were expressed relative to that at zero superficial density. These ratios represent the self-absorption factor for each individual source. The results obtained in this experiment are listed in Table XV.

SELF-ABSORPTION OF C ¹¹ IN BaCO ₃		RADIATION
Source No.	Superficial density mg/cm ²	Relative specific activity
1	3.86	0.899
2	6.64	0.850
3	8.61	0.827
4	9.69	0.804
5	9.96	0.797
6	23.79	0.678

Table XV

In Fig. 40, curve(c) is a graphical representation of the self-absorption versus source superficial density. In the same figure, curve(a) represents the self-absorption of C^{11} radiation in polyethylene as reported by Rosenfeld et al.⁽⁴²⁾, and curve(b) is the same in polystyrene obtained by Crandall et al.⁽³⁹⁾

Semi-empirical equation

In comparing the self-absorption curves of Rosenfeld et al. and of Crandall et al. to that obtained in this work, it was noticed that for the same self-absorption factor the corresponding superficial densities of polystyrene and $BaCO_3$ were at a constant ratio of 2.5:1. This regularity indicated that the self-absorption of the C¹¹ beta radiation depended on the density of the source materials. It was therefore concluded that if any of the existing semi-empirical equations for self-absorption could describe the results of this work, it should also describe the results of Crandall et al., if proper self-absorption coefficients were used in each case.

In the course of some self-absorption studies conducted in this Laboratory, Cooper, Kahana and Yaffe⁽¹¹⁵⁾ derived an equation which described quite accurately the results obtained for measurements in 2π geometry. Since a 4π counter is nothing but two counters of 2π geometry operating in parallel, it was anticipated that the same equation would be applicable in this case too.

This equation was derived on the assumption that an

SELF-ABSORPTION CURVES FOR C¹¹ BETA RADIATION IN VARIOUS MATERIALS

- (a) Polyethylene⁽⁴²⁾
- (b) Polystyrene⁽³⁹⁾
- (c) BaCO₃ (this work)

- Curve obtained by means of the

Cooper, Kahana and Yaffe equation(Curves b & c)

- O Experimental points for the BaCO₃ experiment
- ▲ Experimental points obtained by Crandall et al.⁽³⁹⁾ for polystyrene



exponential absorption law could be applied to the radiation emitted by a volume element dV within the source, which must travel a distance r to reach the surface. It was also assumed that all radiations reaching the surface were measured. The final equation is:

$$A = A_{0}\left(\frac{1 - e^{-\mu x}}{\mu x}\right) + \frac{1}{2}\mu x + \mu x\left(\frac{1}{2}\ln\mu x - \frac{1}{4}\right) - \left(\frac{\mu x}{3}\right)^{2}.....(23)$$

where A = actual activity measured.

 A_0 = activity that would be measured if no

self-absorption were present.

- μ = self-absorption coefficient in cm⁻¹.
- x =thickness of the source in cm.

 $\gamma = 0.5772 = Euler's$ constant which appeared during the integration of their first differential equation.

Since source superficial density was used in this work instead of thickness, x was substituted by its equivalent

$$\mathbf{x} = \frac{\mathbf{d}_{\mathbf{g}}}{\mathbf{d}_{\mathbf{y}}} \qquad (24)$$

where $d_s = superficial$ density of the source.

 d_{v} = its volume density.

Hence Equation (23) became

 $\mathbf{S} = \frac{1 - e^{-Kd_s}}{Kd_s} + 0.0386 \ Kd_s + \frac{1}{2} \ Kd_s \ln Kd_s - \left(\frac{Kd_s}{3}\right)^2 \ \dots \ (25)$

where S is the self-absorption factor $\frac{A}{A_{O}}$.

K is the new density dependent self-absorption coefficient.

$$K = \frac{\mu}{d_v} \qquad (26)$$

0.0386 = $(\frac{1}{2} - \frac{1}{4})$ i.e. the coefficient of the term Kd_g .

Very good agreement was obtained between the calculated and experimental values for both polystyrene and BaCO3.

The solid lines(b) and (c) in Fig. 40 represent the self-absorption, calculated by means of Equation 25, while the points represent the experimentally determined values. The self-absorption coefficients used were:

 $K_{PST} = 0.0054 \text{ cm}^2/\text{mg}.$ $K_{BaCO_3} = 0.0135 \text{ cm}^2/\text{mg} \text{ or } 2.5 \text{ x } K_{PST}.$

This relationship between K_{PST} and K_{BaCO3} shows that the self-absorption is proportional to the density of the source material.

It should be noted here that the above quoted K values did not fit any of the known empirical equations relating the self-absorption coefficient to the maximum energy of the beta spectrum. This may be due to the fact that C^{11} is a positron emitter and therefore has a different beta energy spectrum from the negatron emitters for which these empirical equations have been derived.

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