RESEARCH ARTICLES

J. Sci. Soc. Thailand, **3** (1977), 102-117

FAST NEUTRON BOMBARDMENT OF ^{64}Ni AND DECAY OF ^{64}Co

PRASIT CHAROENKWAN

Department of Physics, Faculty of Science, Chiang Mai University, Chiang Mai, Thailand

(Received 25 July 1977)

Summary

Bombardment of nickel enriched in ^{64}Ni with 14.3-MeV neutrons from the neutron generator produce three activities with the half-lives of 26 ± 1 s, 2.25 ± 0.2 min, and 6 ± 0.5 min. The decays of these activities have been investigated using a plastic scintillator, NaI(Tl) detectors, and a Ge(Li) detector. Beta and gamma ray spectroscopy verifies the existence of ^{63}Co, ^{64}Co, and ^{61}Fe following the ^{64}Ni(n,d)^{63}Co, ^{64}Ni(n,p)^{64}Co, and ^{64}Ni(n,α)^{61}Fe reactions, respectively. The decay schemes of ^{63}Co and ^{61}Fe have been modified and a partial decay scheme of ^{64}Co has been proposed.

Introduction

There have been various conflicting reports from investigations on fast neutron bombardment of ^{64}Ni and the assignment of the isotope ^{64}Co. Parmley *et al.*[1] assigned a 4-5-min activity to ^{64}Co resulting from the ^{64}Ni(n,p)^{64}Co reaction using neutrons produced by 22-MeV deuteron bombardment of Be. Ricci *et al.*[2,3] reported a 6.0 ± 0.5 min beta and gamma ray activity in the radiochemically separated Fe fraction from neutron irradiated Ni and Cu. The maximum beta ray energy was measured to be 2.8 MeV. Preiss and Fink[4] assigned 2.0-min and 7.8-min activities to ^{64m}Co and ^{64g}Co, respectively. Vol'ter *et al.*[5] irradiated a Ni sample enriched in ^{64}Ni with 14.1-MeV neutrons and reported the cross sections for production of three activities. One of these reactions was taken to be ^{64}Ni(n,p)^{64m}Co (2 ± 0.2 min). Strain *et al.*[6] observed a 28 ± 2 s activity which decayed with 95-KeV gamma ray and 3-4 MeV beta ray, and they assigned this activity to ^{64}Co. Grench *et al.*[7] assigned a 6.15-min activity to the decay of ^{61}Fe following the bombardment of 96% enriched ^{64}Ni with 15-19 MeV neutrons. They also observed the beta activity of 2 min or less with an end-point energy of 3.5-4 MeV and with no observable gamma ray associated; the origin of the activity was not determined. The decay scheme of ^{61}Fe was then proposed. Ward *et al.*[8] observed a 0.40-s activity with a beta end-point energy of 7.0 ± 0.5 MeV assigned to ^{64}Co. Also Ward *et al.* assigned a 26 ± 1 s activity as compared to the 28 ± 2 s activity observed by Strain *et al.*[6] with a single gamma ray of 86.3 ± 0.7 KeV and a coincident beta ray of 3.6 ± 0.2 MeV to ^{63}Co.

In the present work attempt is made to investigate what activities are produced following the fast neutron bombardment of ^{64}Ni, to clarify some conflicting reports mentioned above, and to study the decay of ^{64}Co.

Materials and Method

A 20-mg sample of 99% enriched ^{64}Ni was obtained from the Stable Isotope Division of the Oak Ridge National Laboratory, Oak Ridge, Tennessee. The sample was bombarded with 14.3-MeV neutrons from the Chiang Mai Neutron Generator and with fast neutrons from the Thai Research Reactor in Bangkok. After bombardment, the sample was then transported to the detection system.

Gamma ray spectrometry was carried out using the 12.6 cm x 12.6 cm and 7.6 cm x 7.6 cm NaI(Tl) detectors and a 10 mm x 5 mm lithium drifted germanium detector. Because of weak beta activities produced, beta spectrometry was carried out using a 4.5 cm x 2.8 cm cylindrical plastic scintillator. Gross beta and gamma decay measurements were performed using a Nuclear Data 400-Channel Analyzer in the multi-scaler mode. The NaI(Tl) detectors and the plastic scintillator were used in connection with the 200-Channel Packard Analyzer, the 1024-Channel ORTEC Analyzer, and the 512-Channel Nuclear Data Analyzer, while the Ge(Li) detector was used with the 4096-Channel ORTEC Analyzer.

Results

Gammy Ray Spectrum

Initial bombardments were made with 14.3-MeV neutrons of flux in the order of $10^9/cm^2$ s from the neutron generator[9]. The gamma ray spectrum detected by the 7.6 cm x 7.6 cm NaI(Tl) detector from several 1-min bombardments is shown in Fig. 1(a). Fig. 1(b) indicates the decay of the same gamma ray spectrum after an elapsed time of 2 min. The following gamma rays were found to decay with the half-life of 6 ± .5 min: 0.12, 0.30, 0.63, 0.84, 1.03, 1.20, and 1.65 MeV. These gamma rays observed are identified to follow the decay of ^{61}Fe resulting from the $^{64}Ni(n,\alpha)^{61}Fe$ reaction in agreement with the work of Grench *et al.*[7]. Two new gamma rays of 0.63 and 0.84 MeV in energy were not reported by Grench *et al.*, but they were found to decay with the same half-life as ^{61}Fe. As these two gamma rays then fit the decay scheme as proposed by Grench *et al.* in Fig. 2, they are here assigned to be gamma rays following the decay of ^{61}Fe. Furthermore, gamma rays of 0.51, 1.35, and 1.80 MeV in energy were found to decay with the half-life of 2.2 min which is much shorter than the half-life of ^{61}Fe. In the measurements of Grench *et al.*[7],

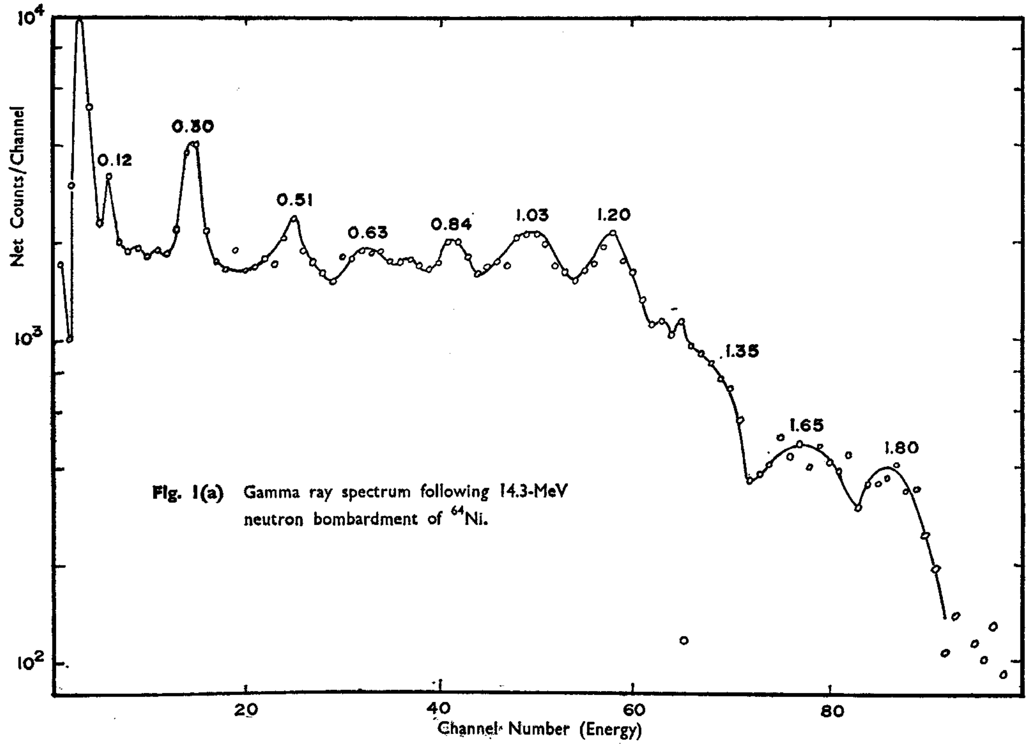

Fig. 1(a) Gamma ray spectrum following 14.3-MeV neutron bombardment of ^{64}Ni.

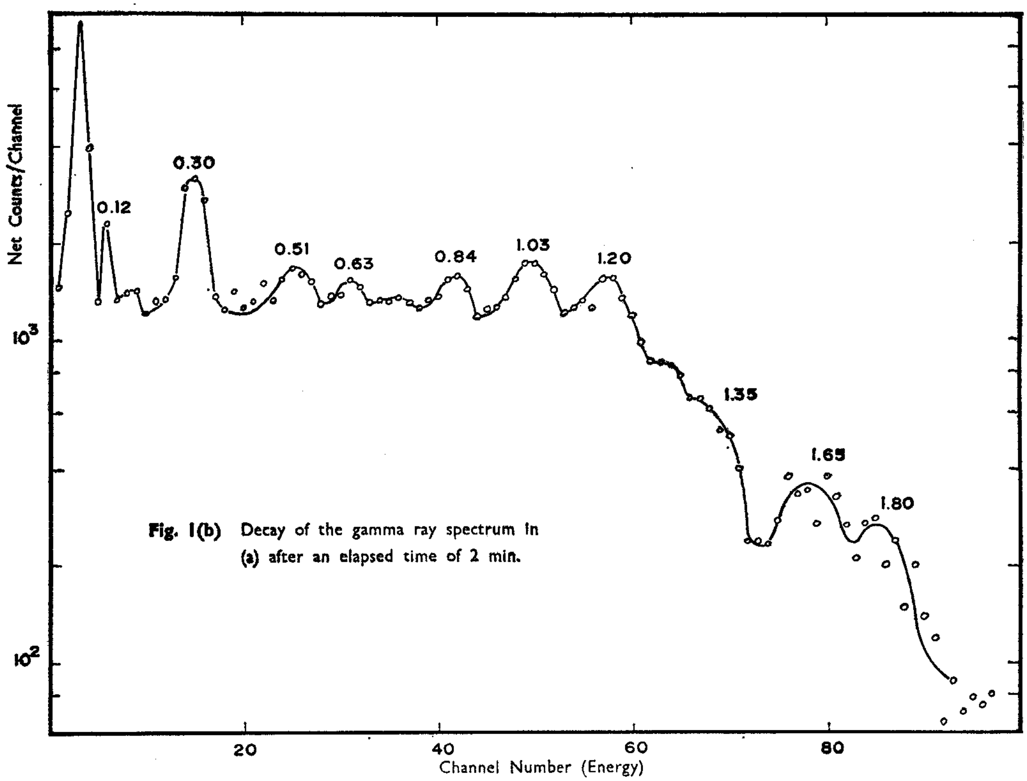

Fig. 1(b) Decay of the gamma ray spectrum in (a) after an elapsed time of 2 min.

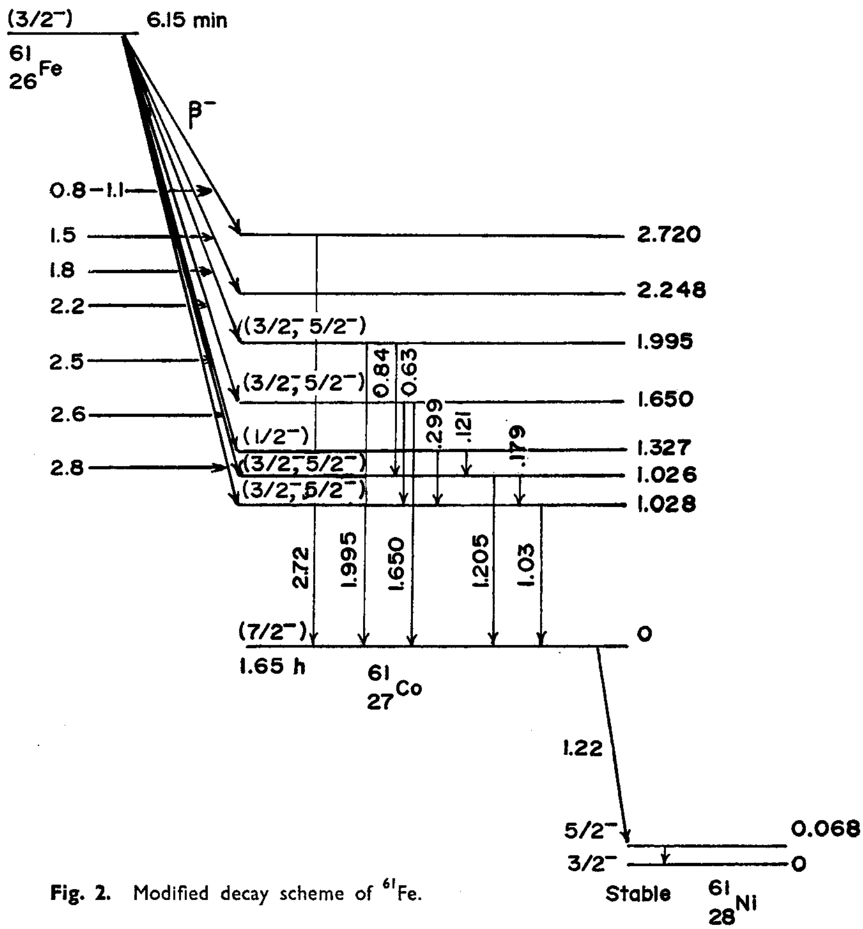

Fig. 2. Modified decay scheme of ^{61}Fe.

they also observed an activity of less intensity, compared to that of ^{61}Fe, with the half-life of 2 min or less. The present investigation then confirms the presence of the 2-min activity as earlier reported by Preiss and Fink[4]. On the basis of the observed gamma ray spectrum and relative Ni isotopic abundances, reactions with the Ni isotopes other than ^{64}Ni were ruled out. On examining the Chart of Nuclides, only the $^{64}Ni(n,\alpha)^{61}Fe$, $^{64}Ni(n,p)^{64}Co$, and $^{64}Ni(n,d)^{63}Co$ reactions are possible. On the basis of the calculations using the empirical mass formula by Seeger[10], the threshold energies for the $^{64}Ni(n,\alpha)^{61}Fe$, $^{64}Ni(n,p)^{64}Co$, and $^{64}Ni(n,d)^{63}Co$ reactions are estimated at 1.2, 5.9, and 9.4 MeV, respectively.

To see what type of gamma ray spectrum could be produced at other energies, the sample sandwiched between the cadmium absorbers to reduce production of the $^{64}Ni(n,\gamma)^{65}Ni$ reaction, was irradiated with fast and thermal neutrons of high flux at the core of the Thai Research Reactor. Gamma ray spectrum analysis was carried out using the 12.6 cm × 12.6 cm NaI(Tl) detector of 7.8% resolution on the 661 KeV gamma ray from the decay of ^{137}Cs and the Ge(Li) detector of 2.2-KeV full width at half maximum on the 1.33-MeV gamma ray from the decay of ^{60}Co. The NaI(Tl) detector was used with the 512-channel analyzer, and the Ge(Li) detector was used with the 4096-channel analyzer.

Gamma ray spectrum of irradiated enriched ^{64}Ni taken with Ge(Li) detector is shown in Fig. 3(a) for the low energy portion and in Fig. 3(b) for the high energy portion. Gamma ray spectrum of 0.368, 0.51, 1.14, 1.49, 1.62, and 1.72 MeV in energy decayed with the half-life of 2.55 h are identified to be gamma ray energies with intensities following the decay of ^{65}Ni. Gamma rays of 0.206 and 1.78 MeV in energy observed in this work were examined to decay with the half-lives of 26 $\pm$ 1 s and 2.25 $\pm$ .2 min, respectively. As the 0.51- and 1.35-MeV gamma rays are of weak intensities compared to those of gamma rays following the decay of ^{65}Ni, the activities of these two gamma rays could not be accurately determined. Apparently, the 0.51- and 1.35-MeV gamma rays were observed to decay with the half-life in the order of minutes. The 2.55-h activity was produced from the $^{64}Ni(n,\gamma)^{65}Ni$ reaction whereas the 26-s and 2.25-min activities were perhaps produced from the $^{64}Ni(n,d)^{63}Co$ and or $^{64}Ni(n,p)^{64}Co$ reactions. The production of ^{65}Ni was possible due to high thermal neutron flux in the reactor. As the irradiation position of ^{64}Ni was moved further away from the core of the reactor, the 0.206- and 1.78-MeV gamma rays disappeared whereas the 0.368-, 0.51-, 1.14-, 1.49-, 1,62-, and 1.72-MeV gamma rays still remained. Half-life analysis of the 1.78-MeV gamma rays agrees with the result of Preiss and Fink[4]. The shorter lived 26-s activity does not give the same gamma ray energy as that of ^{63}Co as reported by Ward *et al.*[8] and is perhaps the new one previously unreported.

As the efficiency of the Ge(Li) detector decreases rapidly at high energy, the gamma ray spectrum following fast and thermal neutron irradiation in the reactor was also measured using the 12.6 cm × 12.6 cm NaI(Tl) detector as shown in Fig. 4. Gamma ray energies following the decay of ^{65}Ni with correct intensities were then

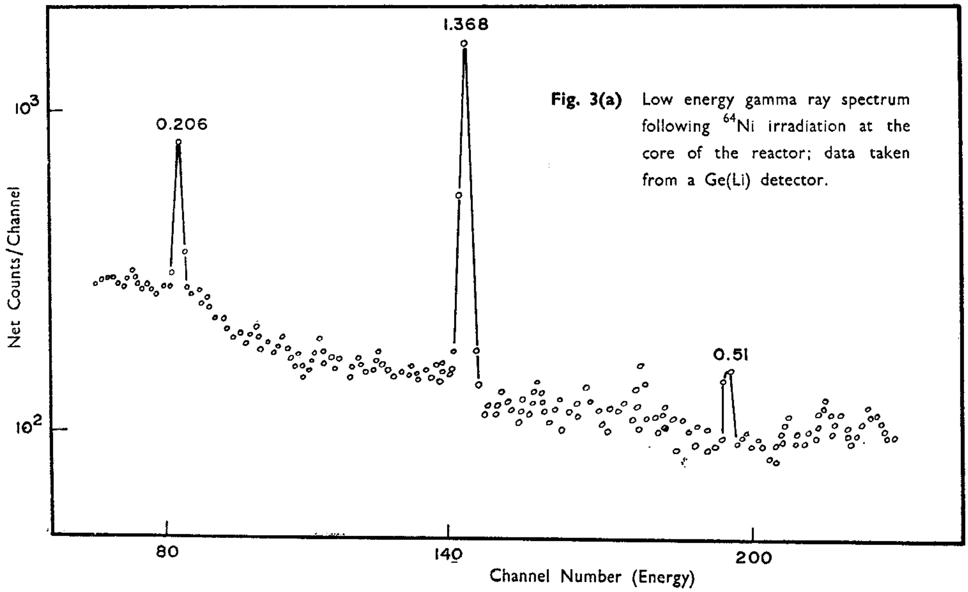

Fig. 3(a) Low energy gamma ray spectrum following ^{64}Ni irradiation at the core of the reactor; data taken from a Ge(Li) detector.

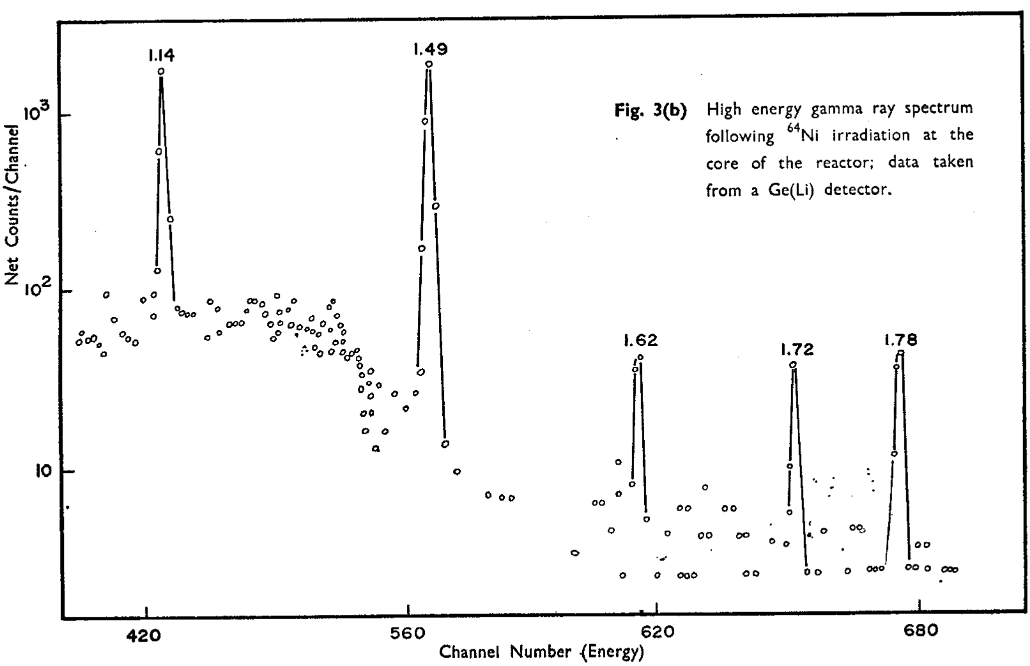

Fig. 3(b) High energy gamma ray spectrum following ^{64}Ni irradiation at the core of the reactor; data taken from a Ge(Li) detector.

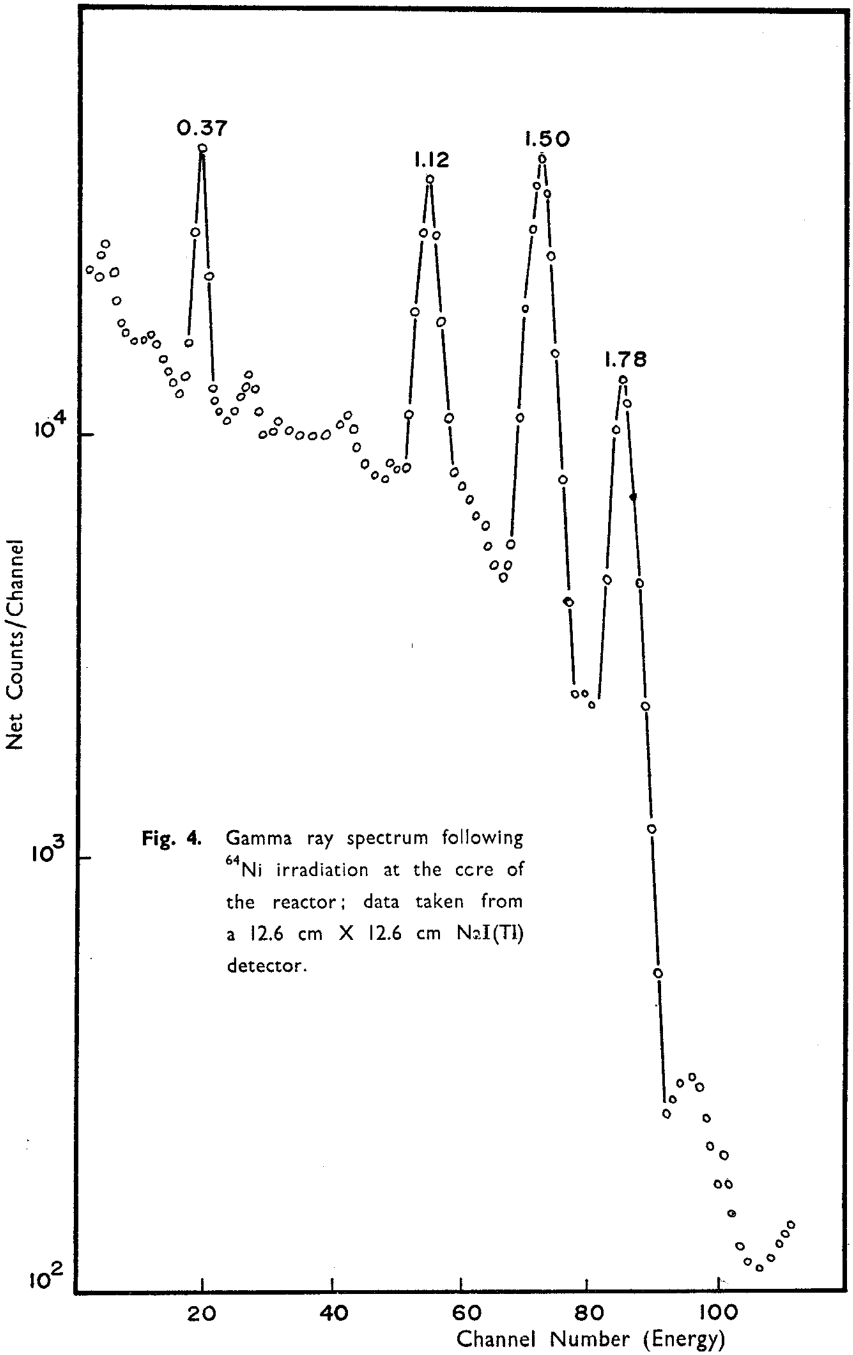

Fig. 4. Gamma ray spectrum following ^{64}Ni irradiation at the ccre of the reactor; data taken from a 12.6 cm X 12.6 cm NaI(Tl) detector.

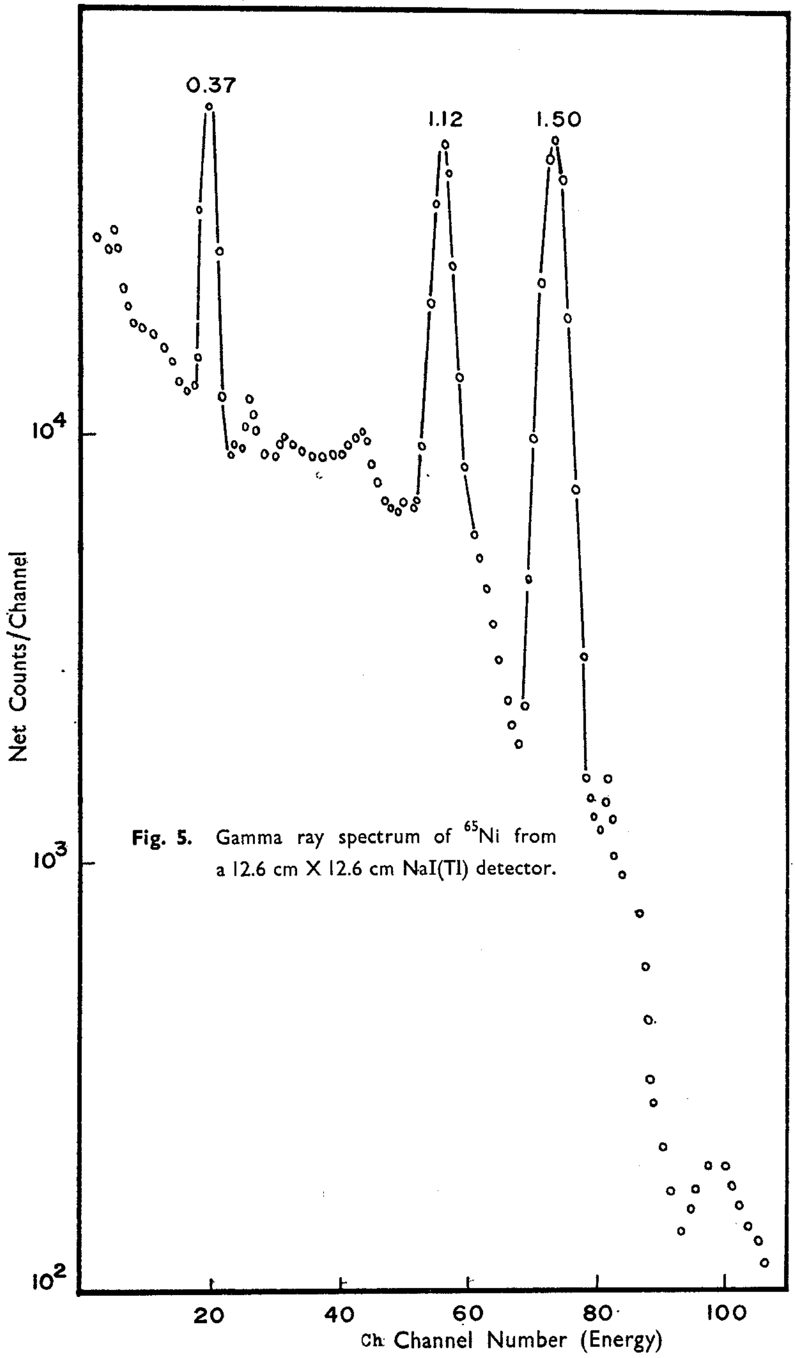

Fig. 5. Gamma ray spectrum of ^{65}Ni from a 12.6 cm X 12.6 cm NaI(Tl) detector.

identified. The gross gamma decay of the 1.78-MeV region was analyzed to have the half-life of 2.25 ± .2 min. Fig. 5 reveals the gamma ray spectrum of ^{65}Ni, decaying with the half-life of 2.55 h after the counting time of 10 min had passed.

Beta Ray Spectrum

The beta ray spectrum emitted following the 14.3-MeV neutron bombardment of ^{64}Ni was investigated using a 4.5 cm x 2.8 cm cylindrical plastic scintillator, a single channel analyzer, and a 1024-channel analyzer. Half-life measurement using the single channel analyzer with the discriminator set up at some certain energy levels reveal the activities tabulated in Table I.

TABLE I: BETA ACTIVITIES

Beta discriminator setting (MeV)	Half-life observed	Activity produced (%)
0.624	18 s 2 min 6 min	35 17 48
1.50	18 s 2 min 6 min	33 21 46
2.00	23 s	–

Table I indicates that the shorter lived beta activities of approximately 23-s half-life were produced and emitted beta rays with an end-point energy above 2 MeV.

^{64}Ni was also irradiated with thermal and fast neutrons at the core of the reactor. Immediately following the irradiation, a multiscaler mode was operated on the gross beta decay at energy above 2 MeV, and the beta activity of 26 s was clearly separated from the 2.55-h beta activity of ^{65}Ni produced by the $^{64}Ni(n,\gamma)^{65}Ni$ reaction. This result agrees with the one obtained from the 14.3-MeV neutron bombardment of ^{64}Ni, and thus confirms the presence of the short lived 26-s activity.

The beta ray spectrum following the bombardment of ^{64}Ni with 14.3-MeV neutrons was detected by the cylindrical plastic scintillator and recorded by the 1024-channel analyzer. Several 1-min bombardments were needed to cover the range of beta spectrum with energy up to 3.5 MeV. The entire beta spectrum was recorded at the following time intervals: 0.30 s, 40-160 s, 170-290 s, and 300-720 s. The Kurie plot of the beta spectrum at these four different time intervals indicates a least four prominent components of allowed shape with maximum end-point energies tabulated in Table II. The Kurie plot of the beta spectrum recorded in the 0-30 s time interval is shown in Fig. 6.

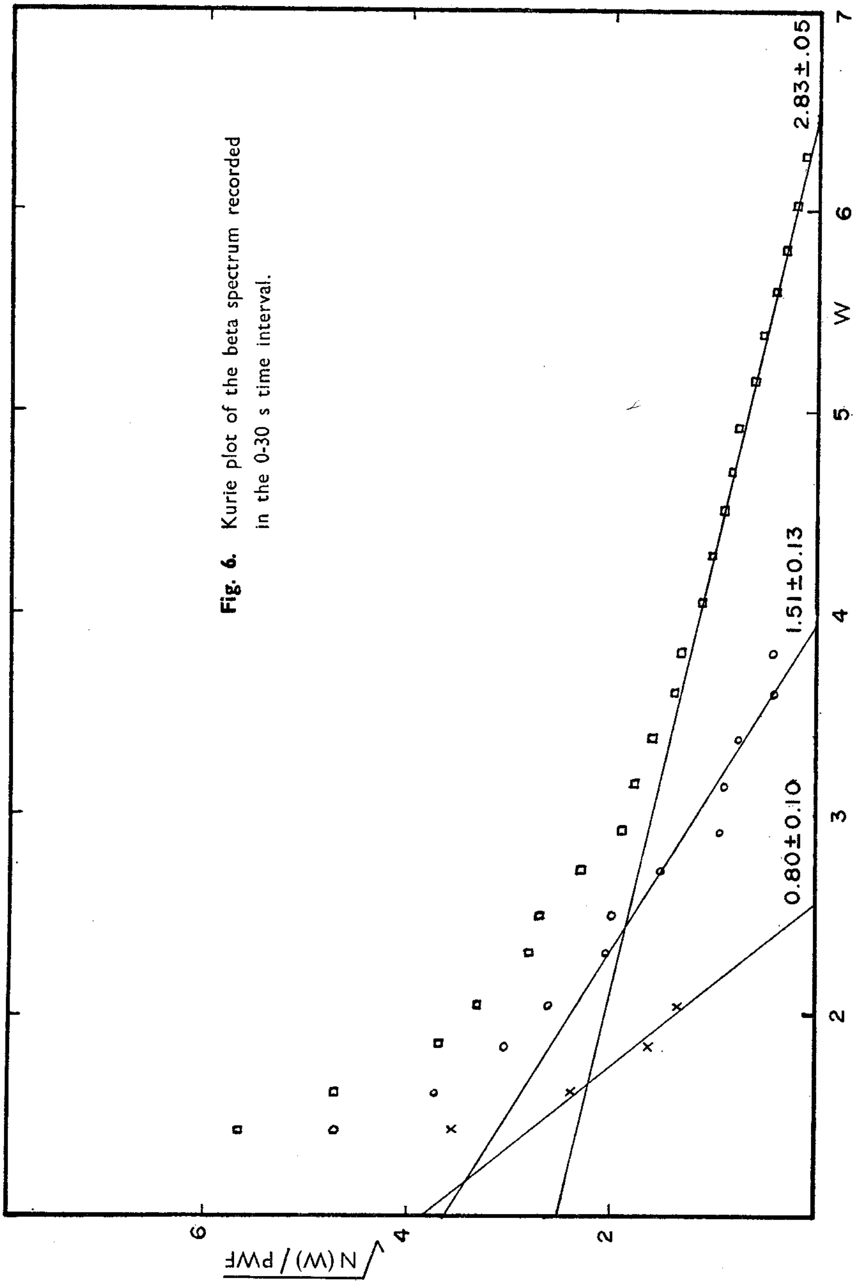

Fig. 6. Kurie plot of the beta spectrum recorded in the 0-30 s time interval.

TABLE II: BETA COMPONENTS FOLLOWING 14.3-MeV NEUTRON BOMBARDMENT OF ^{64}Ni

Time interval of spectrum recorded (s)	End-point energies (MeV)	Relative intensities (%)
0–30	2.83 ± 0.05 1.51 ± 0.13 0.80 ± 0.10	59 31 10
40–160	2.68 ± 0.06 1.51 ± 0.08 0.77 ± 0.09	59 30 11
170–290	2.69 ± 0.05 1.52 ± 0.09 0.87 ± 0.08	54 35 11
300–720	2.66 ± 0.03 1.53 ± 0.08 1.14 ± 0.12	48 38 14

The beta components with end-point energies at 0.80, 1.14, 1.51, and 2.68 MeV mainly decay with the half-life of 6 min and are identified to follow the decay of ^{61}Fe. This measurement gives the 2.68-MeV beta component in agreement with the work of Grench *et al.*[7] and the 1.51-MeV beta component lower than the 1.80-MeV component as reported by Grench *et al.*. The 0.80-MeV beta component was not previously reported in the decay scheme of ^{61}Fe as proposed by Grench *et al.*; however, if existed, it can fit in that decay scheme (Fig. 2). The 2.83-MeV beta component mainly decays with the half-life of 2 min mixed with the 6-min activity of ^{61}Fe. Thus the beta spectrum with the end-point energy of 2.83 MeV and half-life of approximately 2 min decays with the same half-life as the 1.80-MeV gamma ray. As only four beta spectrum displays of diffferent time intervals mentioned in Table 2 following each bombardment could be recorded in the oscilloscope of the multi-channel analyzer, it was not possible to separate the beta spectrum of the 26-s activity with the end-point energy above 2 MeV from the 2- and 6-min activities.

Coincidence Measurements

A standard fast-slow coincidence system was used to provide the beta-gamma coincidence measurement. The resolving time of the fast side of coincidence unit was set to 0.1 μs or less. The 7.6 cm × 7.6 cm NaI(Tl) crystal was used for a gamma detector and the 4.5 cm × 2.8 cm plastic scintillator as a beta detector. To examine gamma rays feeding from the beta transition, the beta discriminator

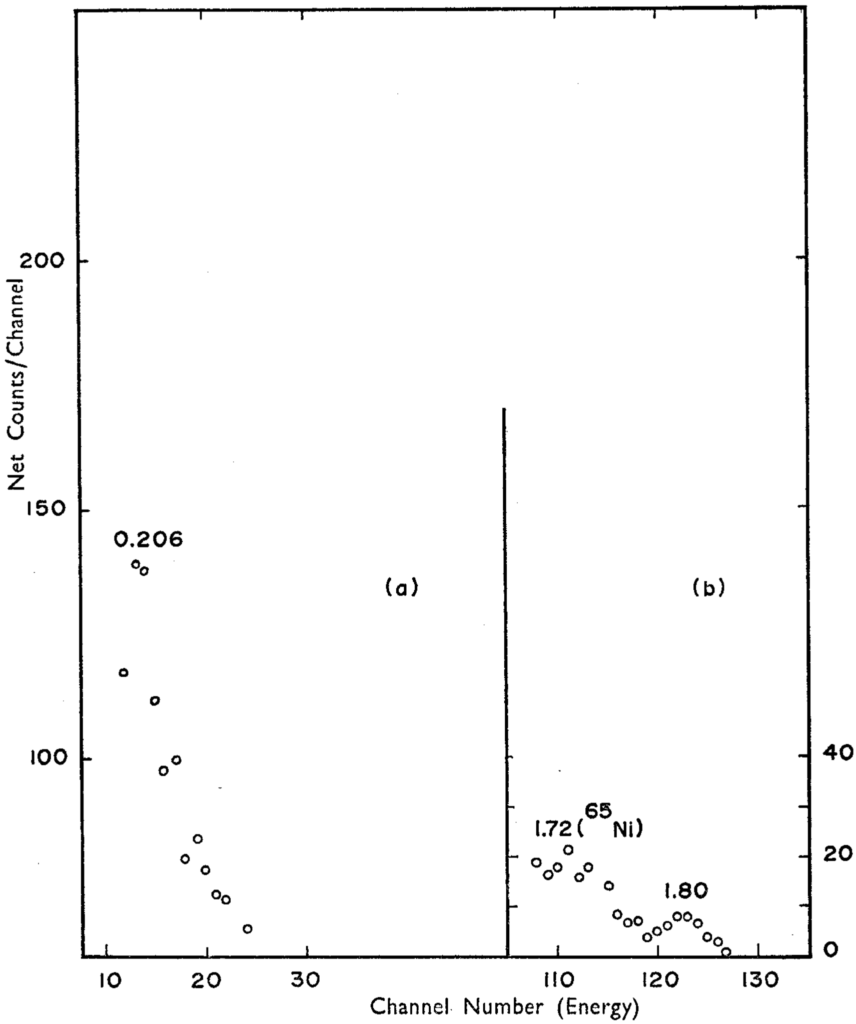

Fig. 7. Gamma ray coincidence spectra. (a) Gamma ray spectrum in coincidence with the 3.50-MeV beta transition. (b) Gamma ray spectrum in coincidence with the 2.83 MeV beta transition.

was successively set to accept beta rays above some certain energy levels such as 2.0 MeV, 3.0 MeV, and other energy levels. All gamma rays in coincidence with the beta spectrum were stored in the 400-channel analyzer. A scaler was used to record data related to the half-life measurement. To assist in writing the proposed decay schemes of the 26-s and 2.25-min activities, the 0.206-Mev gamma ray due to the decay of the 26-s activity was found to be in coincidence with the 3.5-Mev end-point beta component and the 0.51-and 1.80-Mev due to the decay of the 2.25-min activity were found to be in coincidence with the 2.83-MeV end-point beta component as shown in Fig. 7.

Beta-gamma concidences following the decay of the 6-min ^{61}Fe activity were not measured in the present investigation, and the decay scheme modified in Fig. 2 relies on the coincidence data of Grench *et al.*[7] and the beta and gamma ray data in the present investigation.

Discussion

The present measurements confirm the existence of the 6-min activity of ^{61}Fe observed following the 14.3 Mev neutron bombardment of ^{64}Ni in agreement with the work of Grench *et al.*[7]. Two new gamma rays of 0.63 and 0.84 MeV and two new beta components of 0.80 and 1.51-MeV end-point energies of 6-min activity are reported and thus the decay scheme of ^{61}Fe should be modified as shown in Fig. 2.

The presence of the 26-s activity with gamma ray energy of 0.206 MeV and beta ray with an end-point energy above 2 MeV agrees with the work of Ward *et al.*[8] and is here assigned to the existence of ^{63}Co. In the measurement of Ward *et al.*, only a gamma ray of 0.86 MeV in energy which decays with the activity of 26 s was reported, not the 0.206-MeV gamma ray. This absence in the work of Ward *et al.* can be explained from the low 14.8-MeV neutron flux while the fast neutron flux from the reactor used in this measurement is much higher. As the 0.206-MeV gamma ray is in coincidence with the 3.5-MeV end-point beta component, the decay scheme of ^{63}Co is modified in Fig. 8.

The existence of the 2.25-min activity following the fast neutron bombardment of ^{64}Ni is here assigned to ^{64}Co after the identification of ^{61}Fe and ^{63}Co. The 2.25-min activity was observed to have a beta end-point energy of 2.83 MeV with log ft value of 4.7 and gamma rays of 0.51, 1.35, and 1.78 MeV. The presence of the 1.36-MeV gamma ray is in agreement with the adopted level scheme[11] in ^{64}Ni and perhaps this gamma ray is fed from the first 2^+ excited level to the ground level of 0^+. As the measurement of Ward *et al.* reported the 0.40-s activity observed to have a beta end-point energy of 7.0±0.50 MeV without the presence of the gamma ray of the same activity, the 2.25-min activity found from the present measurement should be assigned to an isomer in ^{64}Co. From shell model considerations, the 27th proton and the 37th neutron could couple giving a resultant spin of 1^+ for the lower spin level and 4^+ or 5^+ for the higher spin level. A transition between the very high spin level and the very low spin level requires a large spin change leading to high order multipole transition which has a long lifetime; that is, nuclear isomer exists.

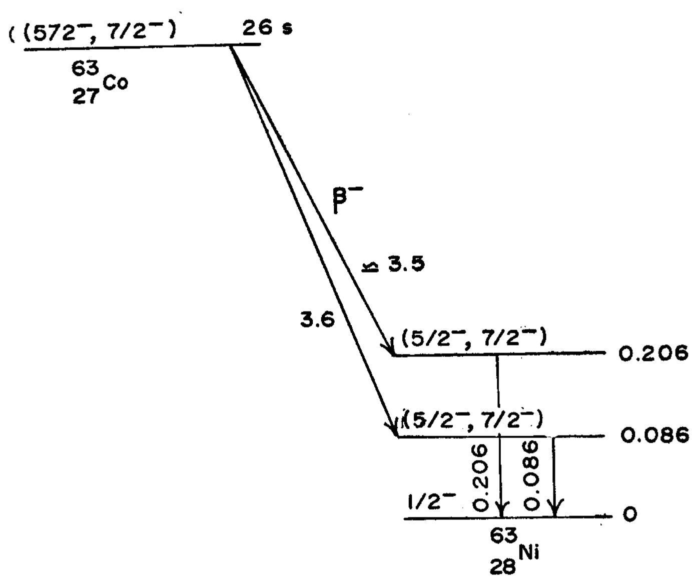

Fig. 8. Proposed decay scheme of ^{63}Co.

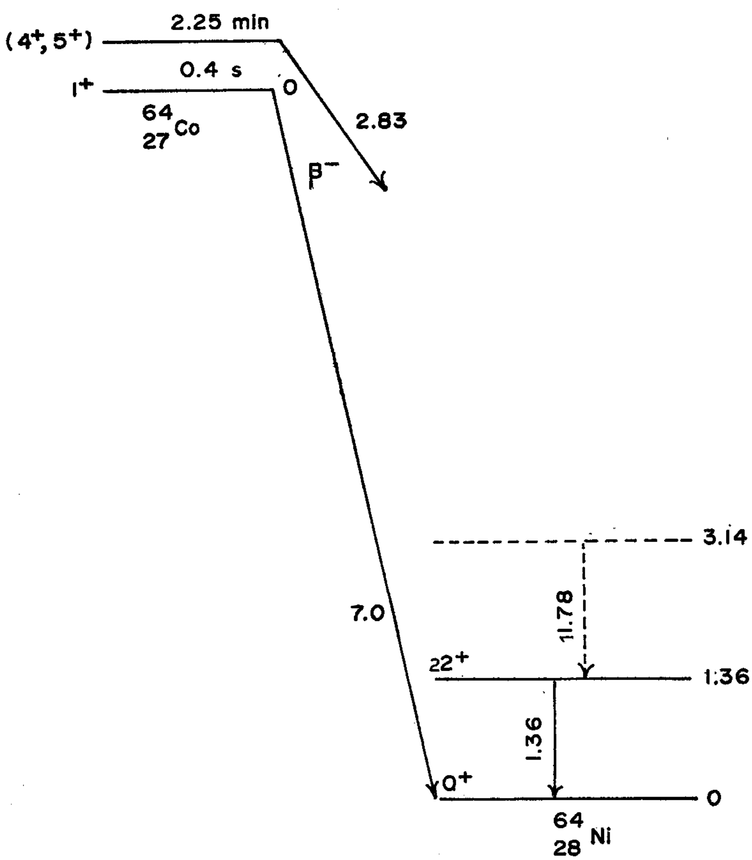

Fig. 9. Proposed decay scheme of ^{64}Co.

It should be pointed out that most of the even mass number Co nuclides are known to have isomeric states with spin and parity sequence of 5^+ and 2^+. Since the work of Ward *et al.*[8] assigned spin of 1^+ to the ground level in ^{64}Co decaying to the ground level in ^{64}Ni with the beta transition of 7.0±0.5 MeV, the present investigation assigns the 2.25-min isomer with high spin possibly 4^+ or 5^+. Since the 7.0-MeV end-point beta transition feeds the ground level, the 2.83-MeV end-point beta transition should feed the much higher excited level. Coincidence data in Fig. 7 confirm this assignment and a partial decay scheme of ^{64}Co is proposed in Fig. 9.

Acknowledgements. The author wishes to thank graduate students Krayim Santrakul, Charnnarong Tampanitwat, and Vivat Teeyasoontranont for bombarding the sample and assisting in some part of the experiment. Also the assistance of the staff members of the Thai Research Reactor in Bangkok for irradiating the sample and setting up the detection equipment is acknowledged.

This work was supported in part by the National Research Council.

References

1. Parmley, T.J., Moyer, B.J., and Lilly, R.C. (1949) *Phys. Rev.* **75**, 619.
2. Ricci, E., Pahissa Campa, J., and Massis, N. (1955) *Z. Naturforsch.* **10** *a*, 654.
3. Ricci, E., Pahissa Campa, J., and Nassis N. (1957) *Publ. Com. Nac. Energ. At. Argent.* **1**, 129.
4. Preiss, I.L. and Fink, R.W. (1960) *Nucl. Phys.* **15**, 326.
5. Vol' ter., A.K, Gonchar, V.Y., Zalyubovskii II., Latyshev, G.D., and Coursin, G.P. (1962) *Izv. Akad. Nauk. SSSR, Ser. Fiz.* **26**, 1079.
6. Strain, J.E. and Ross, W.J. (1966) *J. Inorg. Nucl. Chem.* **28**, 2075.
7. Grench, H.A., Coop, K.L., and Menlove, H.A. (1967) *Phys. Rev.* **161**, 1118.
8. Ward, T.E., Pile, P.H., and Kuroda, P.K. T (1969) *J. Inorg. Nucl. Chem.* **31**, 2679.
9. Charoenkwan, P. and Vilaithong, T (1976) *J. Sci. Fac., Chiang Mai Univ.* **2**, 27.
10. Seeger, P.A. (1961) *Nucl. Phys.* **25**, 37.
11. Horen, D.J., ed. (1973) in *Nuclear Level Schemes*, Academic Press Inc., New york.

บทคัดย่อ

การยิง ^{64}Ni ด้วยนิวตรอนพลังงาน 14.3 MeV จากเครื่องกำเนิดนิวตรอนนั้น ผลิตนิวไคลด์รังสีที่มีครึ่งอายุ 26±1 วินาที 2.25±.2 นาที และ 6±.5 นาที การศึกษาวิธีการสลายตัวของนิวไคลด์รังสีเหล่านี้กระทำได้ โดยใช้หัววัดแบบปลาสติก แบบโซเดียมไอโอไดด์ผสมทาลเลียม และแบบเจอร์มันเนียมลิทเทียม การวัดทางด้านสเปคโตรตสโคปีของรังสีแกมมาและเบตายืนยันการเกิดนิวไคลด์ ^{63}Co ^{64}Co และ ^{61}Fe อันเป็นผลจากปฏิกิริยานิวเคลียร์ ^{64}Ni (n,d)^{63}Co ^{64}Ni (n,p) ^{64}Co และ ^{64}Ni (n,α) ^{61}Fe ตามลำดับ ข้อมูลที่ได้จากการค้นคว้าวิจัยครั้งนี้สามารถนำไปใช้ปรับปรุงแผนภาพการสลายตัวของ ^{63}Co และ ^{64}Fe และเสนอแผนภาพการสลายตัวของ ^{64}Co ได้เป็นครั้งแรก