CHAPTER VI

RARE TYPES OF FISSION

6.1 INTRODUCTION :

6.1a Ternary Fission :

Besides binary fission in which the excited nucleus splits into two fragments of comparable mass, there are also other types of fission of excited nuclei. It was R. Present[1] who on the basis of liquid drop model of Bohr and Wheeler[2] predicted the possibility of tripartition or ternary fission i.e., the division of an excited nucleus into three nuclei of comparable mass. This process is comparatively rare and the evidence for this is meager. L. Rosen and A.M. Hudson (1949) in a preliminary report on the tripartition of U^{235} bombarded with slow neutron, found about 4.3 ternary fission per 10^6 binary fission. In other work on ternary fission, two massive fragments have been observed along with the third particle, usually a long range alpha or in a few cases somewhat heavier than alpha. K.W. Allan and J.T. Dewan (1950) in their investigations of ternary fission of U^{233}, U^{235} and P^{239} found long range alpha particles to be associated with 2 p.c. of fission. The average K.E. of alpha E_α = 15 Mev and E_{max} = 29 Mev.

In an investigation of ternary fission of U^{235} by slow neutron, E.W. Titerton, using photographic emulsion technique observed that most of the long range alpha particles were emitted at right angles to the line of motion of the two massive fission fragments. The distribution of energy among these alpha particles showed a maximum of about 29 Mev with the greater number having energies in the neighbourhood of 15 Mev, corresponding range is about 100 microns. The observed maximum alpha particle energy suggests the idea that the alpha particle is left between the massive fission fragments at the instant of fission and aquires its kinetic energy from the electrostatic fields of the nuclei. He found that the frequency of occurance of ternary fission is about 1 per 400 binary fission events in approximate agreement with the results of Allen and Dewan. He also found a few very short tracks that seemed to indicate the formation of nuclei with charge $z > 2$, but found no evidence of ternary fission into approximately three equal masses.

Experimental evidence of such a process has been sought by different workers[3-7] using a variety of methods, such as, (1) nuclear emulsions impregnated with fissionable nuclides, (2) multiple ionisation chambers (triple coincidence between individual detectors), (3) radio chemical analysis, (4) mica detectors and other types of detectors used in heavy ion bombardments.

From their experimental findings it is found that in three fragment fissions mostly alpha is emitted along with two fragments of conventional type. M.Dakoshski[8-9] et al also reported that emission of short range alpha particle with an energy of 7.7 Mev and with a frequency of one particle per 478 ± 24 fission. The emission was observed predominantly at right angle to the direction of heavy fragment[10]. Bagadanov[11] et al taking into account of alpha recoil effect, concluded that alpha particles are formed preferentially from the light fragment. Nifenecker[12] concluded from the studies of charge distribution in ternary fission of Cf^{252} that alpha particle is formed at the expense of both the fragments. However, all measurement agree that the most probable angle of emission[13] of alpha particle is not quite at right angles to the fragment direction, but it is about 20^{o} closer to the direction of the light fragment than to the heavy. The heavier fragment has largest z and therefore repel the alpha particle more effectively than the light fragment does. The most probable angle of alpha particle emission with respect to the light fragment in the fission of Cf^{252} has been measured to be about 81^{o}.

The relative yield of different particles emitted as third particle in ternary fission has been the subject of interest for different workers throughout the world. But the findings of Muga[14] and his co-workers deserve special mention, when they found that the uranium and plutonium nuclei fissioned with slow neutron break up on rare occasion in the three fragments of comparable masses. The specific heavy fragments emission like that of Na^{24} and Mg^{28} have been observed by Cobble[15-16] and his workers at excitation energy about 20 Mev. Such particles are, therefore, not expected to be produced in sponteneous and slow neutron fission. However, induced fission of high energy nuclear interaction seems to be fertile in this case.

6.1b Probable Explanation of Ternary Fission :

The study of this type of fission faces considerable difficulties as there is no comprehensive theory which can match the various experimental observations. Getting into the problem one can divide the whole process into two stages, viz., (i) that which just proceeds the physical separation of the fragments and (ii) that which actually follows. The division is well reasoned, because after scission the two or more fragments of comparable masses separate under the influence of coulomb field of each other. However, the two different mechanisms[17] of ternary fission production in heavy nuclei are found to be possible. One of them being true ternary fission where an initially strong excited nucleus leads to the formation of a configuration with two necks and the nucleus splits into three fragments within a time on the of 10^{-21} sec. In second mechanism, the heavy fragments with sufficiently large values of z^2/A, may be produced as a result of ordinary binary fission of initial nucleus. If the excitation energy of such fragment exceeds the fission barrier, then after sometimes, it can split in turn into two. As a result of this initial compound nucleus turns out to be split into three fragments. It must be emphasised that in the fission of the heavy fragment, we deal with ordinary binary fission at large excitation energies and generally they are termed as cascade fission. The cross-section for cascade fission increases sharply with increasing z^2/A of the compound nucleus. Also with the increase of excitation energy it increases rapidly.

However, a close comparision of binary fission with that of the ternary fission reveal that both the types of fission have all essential characteristic same except that in energetics. In earlier cases it seems that when alpha particle is emitted, the reduction of the mean kinetic energies of the fragments is greater than that of their mean excitation energy. The mass distribution in both the cases of fission is strikingly similar[18,19]. It leads to the idea that ternary fission is actually a binary fission but it gets over enthusiastic at the moment of scission.

6.1c Quaternary Fission :

Apart from these two types of fission, asymmetric division of the excited nucleus, in the process of de-excitation, into four segments, is also possible, and is known as quaternary fission. Tsin-san Tsaing[3] first observed the possibility of quaternary fission in uranium using emulsion technique in 1947. Very recently Kapoor[20] et al found evidence for the simulteneous emission of two light charged particles in coincidence with fission fragments in thermal neutron induced fission of U^{235}. The extensive experimental results in the case of sponteneous fission[21] of Cf^{252} have shown that the two light charged particle coincidences are of the type He^4-He^4 . He^3-He^4. He^1-He^4, etc. and about 80 p.c. of the coincidence are found to be of the type He^4-He^4.

6.1d Mechanism of Quaternary Fission :

There are number of probable mechanism as to the production of these alpha particles. They are -

(1) The two alpha particles are produced as a result of break up of an unstable Be^8 nucleus emitted in ternary fission. The pairs of alpha particle will be confined to an angular separation of about 4^o.

(2) The two alpha particles are emitted at the same time around the neck region of the excited nucleus. The condition implies a process in which the two alpha particles are left in the neck region by the retracting nuclear matter.

(3) The two alpha particles are emitted independently in the neck region so that the time of emission of the two alpha particles are different. This condition implies an evaporation type of emission from the fragment nuclei.

(4) The two alpha particles are liberated on the same side of the axis of fragment separation, independently of each other, so that the time of emission of the two alpha particles are different. This process requires liberation of the alpha particles from the tips of the deformed fission fragments before they are accelerated significantly. Here the major axis of these fragments do not coincide with the axis of separation. However, the hypothesis which explains all

the observed experimental findings implies that the two alpha particles are being emitted independently from the two accelerating fission fragments within about 10^{-21} seconds after scission, before they have aquired an appreciable part of final observed kinetic energies. It is also found necessary to assume that predominantly the two alphas are emitted on the same side of separation axis.

The experimental results on the energy distribution, angular correlation and relative abundances of various particles in quaternary fission and ternary fission indicate that the two process have very much in common. Although the emission of light charged particle in quaternary fission is similar to the emission mechanism of LCP in ternary fission, yet the mean energies for this particle in quaternary fission are lower than in ternary fission.

Here in our studies on rare types of fission use has been made of emulsion technique as detector. It is believed that the excited nucleus (silver and bromine) left after the cascade process, while in the process of de-excitation by evaporation of nuclear particle may give rise to this type of fission apart from usual binary break up. Overall experimental findings on rare type fission is small and it is still smaller in the case of medium weight nuclei and there is no report of it being studied in the light of quaternary fission. Here an attempt has been made to observe such events with similar characteristics as observed by other workers using varied method of detection. A micro-photograph of the events both ternary and quaternary are shown in the Plate No. 1.

6.2 EXPERIMENTAL PROCEDURE AND SELECTION CRITERIA :

Both the stacks (sample A and sample B) have been used. Preliminary area scanning was done under low magnification (X15) to record all the disintegration stars with $N_h \geqslant 8$. Stars with smaller N_h value were excluded in order to ensure the inclusion of silver and bromine disintegration only. Each star so obtained was then carefully examined under high magnification (2000 , oil immersion objective) to collect the disintegration stars with three or more short tracks having ranges upto 40 microns.

Now the combination of any two or more short tracks associated with primarydisintegration centre may simulate a true binary, ternary or quaternary fission events.

However, for fission events these short tracks should have characteristics of heavy ion as discussed in the chapter V, on binary fission.

Further, for the ternary events, either (1) all the three tracks must have similar track characteristics (i.e., residual range, ionisation, tapering, etc.). This will select events with three fragments of comparable mass. Or (2) two prongs may have more or less similar track characteristics whereas the third one may be somewhat different. This criterion selects events containing two more or less identical fragments while the third a different one.

For quarternary fission of the four short prongs associated with primary disintegration stars we select either (1) star containing a pair of short tracks (R = 10 microns) and a pair of long tracks (R = 50 microns). This selects quaternary fission into pair of light and a pair of heavy nuclei, or (2) we select three short tracks of more or less equal length and fourth one being comparatively longer or shorter. This selects events with three fragments of similar size while the other one is different. It may be pointed out that out of the total of 3400 primary disintegration scrutinised in the sample B, we did not find a single event satisfying the above selection criteria. Only three events were at first recorded to be ternary fission events. However on final analysis they had to be rejected. The results presented below therefore are collected from the sample A only.

6.3 RESULTS OF ANALYSIS AND DISCUSSIONS :

Total number of primary disintegration stars scanned	30,000
Number of binary fission events observed	225
Number of ternary fission events observed	18
Number of quaternary fission events observed	1
Frequency of ternary expressed as a fraction of binary events ...	0.08

Frequency of quaternary expressed as a fraction of binary events	...	0.005
Cross-section for binary	...	0.8 mb

Distribution of heavily ionising prongs (N_h) in case of ternary fission is shown in the fig. 6.1. The mean excitation energies of the ternary and the lone quaternary events have also been calculated from their N_h values and the values obtained are found to be 420 Mev for ternary and 540 Mev for quaternary fission events respectively.

Figures 6.2 and 6.3 give the distribution of ranges and range ratio of the prongs of the ternary events. From the distribution of ranges, we find that their ranges vary from 5 to 40 microns with a maximum number having ranges between 5 to 10 microns, a close approximation to the ranges of binary fission events. Further from the range ratio we find that in most of the cases it is confined to a value within 2 (two), a fact that is observed in binary fission. From the angular distribution (as shown in fig. 6.4) we find that the angle between the prongs may vary over a wide range, such as, extending from 40° when the fragment of the pair are confined to a small divergent cone to 180°, when they are emitted exactly in opposite directions. A broad distribution appears between 100 to 140 together with a isolated peak between 80° to 90°. This isolated peak supports the view that in ternary fission frequency of alpha emission is appreciable and most of them are emitted[13] within 80°-90° angle with respect to the light fragments.

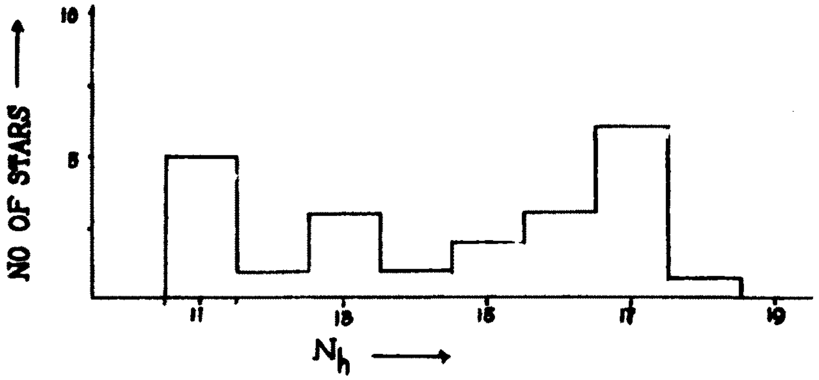


Fig 6.1 N_h DISTRIBUTIONS OF TERNARY FISSION EVENTS IN K^- INTERACTIONS (18 EVENTS)

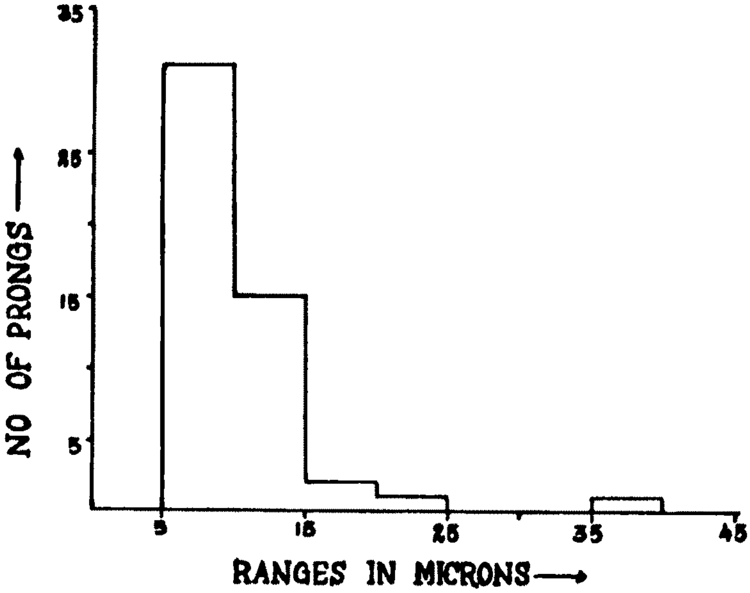


Fig 6.2 RANGE DISTRIBUTION OF THE PRONGS OF THE TERNARY FISSION EVENTS. (18 EVENTS).

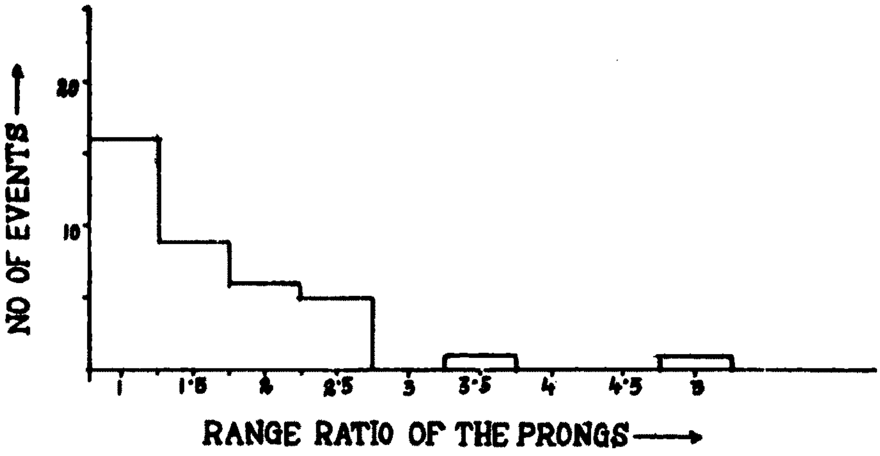


Fig 6.3 RANGE RATIO DISTRIBUTION OF THE PRONGS OF THE TERNARY FISSION EVENTS (18 EVENTS)

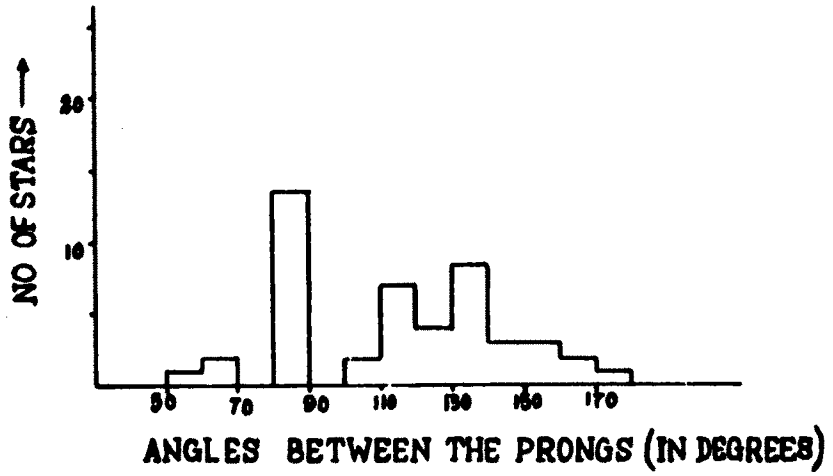


Fig 6.4 ANGULAR DISTRIBUTION BETWEEN THE PRONGS OF THE TERNARY FISSION EVENTS IN K^- INTERACTION. (18 EVENTS)

6.4 ANALYSIS OF A FEW TERNARY EVENTS :

Further an attempt has been made to analyse in order to give a possible identification for a few ternary and the quaternary fission events.

Ternary Events :

The two ternary events where the possible identification could be given are shown in the microphotographs in plate I . The data obtained from the measurements on these events are given in table 6.1.

Event I : An example of fission into comparable masses.

The number of heavily ionising prongs emitted from the disintegrating centre is 16. This corresponds to our excitation energy of $\sim$ 550 Mev. The charge of the fissioning nucleus estimated, as done in earlier chapters comes out to be 17 as a lower limit when interaction in bromine is concerned.

Of the three fragments, for the fission events, one after traversing a range of 55 microns ends in a hammer which is a case with Li^8 fragments

$$Li^8 \xrightarrow{e^-} {}^{*}Be^8 \longrightarrow 2He^4$$

For the other two tracks the possible identities are attributed in such a way that (1) the total V.E.R. for this process does not differ very much from that binary (45 Mev), and (2) a balance of momentum is obtained between these emitted fragments. On the basis of this, it is found that the tracks with range 12 microns may be due to O^{16} and that with 22 microns may be due to C^{12}. It may be pointed out that if we consider the interaction to be in silver than the possible identities for these short tracks will result in large V.E.R. and also balance of momentum can not be obtained. Hence, the event may be a true ternary fission of ${}_{17}Cl^{36}$ nucleus into three fragments of comparable mass, such as,

$$ {}_{17}Cl^{36} \longrightarrow {}_{8}O^{16} + {}_{6}C^{12} + {}_{3}Li^{8} $$

The angular separation between O^{16} and Li^{8} fragments is higher than that of between Li^{8} and C^{12}. It may be due to the fact that the coulombic repulsion between O^{16} and Li^{8} is higher than that between C^{12} and Li^{8}.

Events 2 : A possible example of cascade fission .

Excitation energy and lower limit of charge of pre-fissioning nucleus estimated as in the previous case comes out to be 420 Mev and 24 respectively.

From the microphotograph of the event in plate 1 represented schematically, it is found that the two tracks of ranges 8 and 9 microns are collinear and the third one is emitted almost at right angles (differing by $\sim 12^{o}$) to them. Now from the discussions[8-10] already reported in introduction we may analyse this event on the supposition that this may be a ternary fission event emitting a short range alpha in a direction almost normal to the heavier fragments which repel each other in opposite directions.

Therefore the track of 11 microns may be due to an alpha of energy 3 Mev. The possible identities for the other tracks are given in such a way that (1) the particle traversing the range of 9 microns with slightly of higher charge than that traversing 8 microns, so as to account for the observed deviations of the alpha track from the normal direction and (II) the total V.E.R. obtained does not differ much from that binary process.

In the light of above discussions we find the possible identities to be Ne^{20} and Mg^{24} for the particles traversing 8 and 9 microns respectively. So the possible fission scheme may be given by,

$$_{24}Cr^{48} \longrightarrow {}_{12}Mg^{24} + {}_{10}Ne^{20} + {}_{2}He^{4}$$

This may be a case of cascade fission in which Cr^{48} at first breaks up into two Mg^{24} fragments in a binary process. Of these two fragments one might have exceeded the fission barrier and eventually emitted one alpha which finds itself in between repelling Mg^{24} and Ne^{20}. This alpha moves normally to the direction of repelling Mg^{24} and Ne^{20} fragments. As charge of Mg^{24} is higher than that of Ne^{20}, so the coulombic repulsion between Mg^{24} and alpha is slightly greater than that between alpha and Ne^{20}. Hence the alpha particle is deviated from the normal direction.

Table 6.1 : Data from ternary events.

Events	Primary tracks N_h	Prefission charge (Lower limit)	Tracks	Possible identity	Ranges in Micron	Space angle between	Kinetic energy in Mev	Possible fission scheme
1	$(13+3F)k^-$	Z 17	1	O^{16}	12	90^o	20	$_{17}Cl^{36} \longrightarrow {}_{8}O^{16} + {}_{6}C^{12} + {}_{3}Li^{8}$
			2	C^{12}	22	124^o	25	
			3	Li^{8}	55		20	
							65	
2	$(10+3F)k^-$	Z 24	1	Ne^{20}	8	78^o	26	$_{24}Cr^{48} \longrightarrow {}_{12}Mg^{24} + {}_{10}Ne^{20} + {}_{2}He^{4}$
			2	Mg^{24}	9	102^o	20	
			3	He^{4}	11		3	
							49	

6.5 Quaternary Fission :

A microphotograph of the single event as we have found is shown in the plate no. 1 with a schematic representation. This is similar to that obtained by Chinese worker in 1947. Necessary data obtained from the measurements are furnished in the table 6.2.

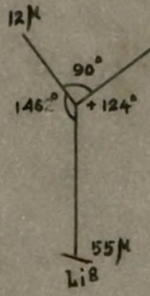
The number of heavily ionising prongs emitted from the disintegrating centre is found to be 17. This corresponds to an excitation energy of about 540 Mev. The charge of the fissioning nucleus as estimated from the identification of the heavily ionising tracks comes out to be 20 as lower limit. The track 1 and 2 having ranges 25 microns each appears to be thin as compared to the track number 3 and 4. Now this event may be a possible case of quaternary fission with two alphas emitted in the same side of the fission axis as discussed in the section 6.1d above.

As in the case of ternary fission we suppose that the V.E.R. in this case also does not differ much from that of binary and ternary fission.

Now of all the possible identities attributed to the tracks 3 and 4 it is found that if we consider both of them to be due to O^{16} then only we get a reasonable V.E.R. This being the case, this event may be a quaternary fission of Ca^{40} which may be represented in the following scheme

$$_{20}Ca^{40} \longrightarrow {_8}O^{16} + {_8}O^{16} + {_2}He^4 + {_2}He^4$$

TERNARY FISSION EVENT NO.1.
(SCHEMATIC REPRESENTATION)

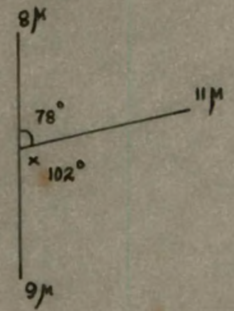


$Cl^{36} \longrightarrow O^{16} + C^{12} + Li^{8}$

V.E.R = 65 MEV.

Fig 6.5

TERNARY FISSION EVENT NO.2.
(SCHEMATIC REPRESENTATION)

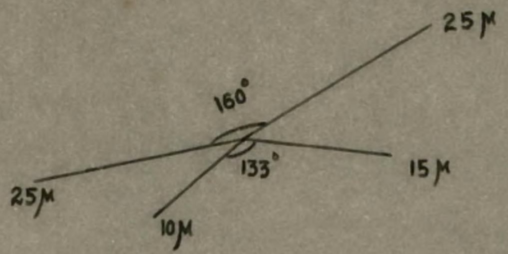


$Cr^{48} \longrightarrow Mg^{24} + Ne^{20} + He^{4}$

V.E.R = 49 MEV.

Fig 6.6

QUATERNARY FISSION EVENT NO.3
(SCHEMATIC REPRESENTATION)



$Ca^{40} \longrightarrow 2O^{16} + 2He^{4}$

V.E.R $\simeq$ 58 MEV.

Fig 6.7

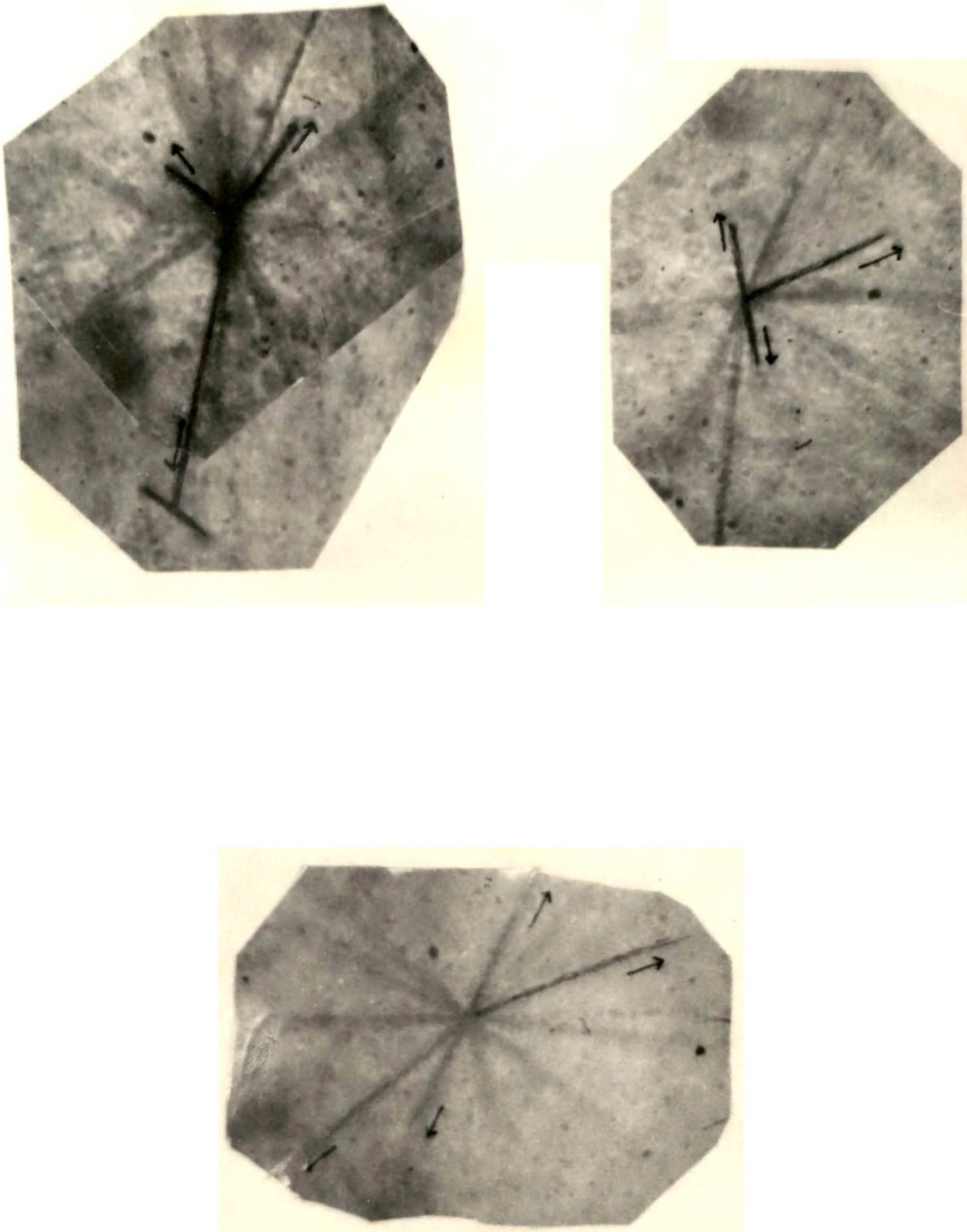


PLATE - 1

The mechanism of formation may be explained in conformity with the discussion given in 6.1d. The excited pre-fissioning Ca^{40} nucleus may at first split into two parts (Ne^{20} each) which at no time ($\sim 10^{-21}$ sec) after scission eject alphas from their deformed tips. The two alphas are found to have same kinetic energies. It is perhaps due to the fact that initial splitting of Ca^{40} is symmetric. An amount of unbalanced momentum appears along the forward direction of heavy fragments. This may be due to fission in a moving system.

Table 6.2 : Data from quaternary events.

Primary	Prefission charges (lower limit)	Tracks	Ranges in micron	Space angle between the tracks	Possible identity	Kinetic energy in Mev	Possible fission scheme
$(13+4F)k^-$	Z = 20	1	25	160°	He^4	6	
		2	25	38°	He^4	6	$Ca^{40} \longrightarrow 2O^{16} + 2He^4$
		3	15	133°	O^{16}	26	
		4	10		O^{16}	20	

						Total = 58	

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