



Introduction

Our choice for the Classics section is the paper by Chatterjee and Sarkar, describing their measurement of the lifetime for uranium to undergo spontaneous fission. It is an outstanding achievement of experimental physics in India, as clear from Amit Roy's article on the life and work of Shyamadas Chatterjee. But there are many more interesting aspects to this paper. We have reproduced the paper as it appeared in 1944 in the journal, *Science and Culture*, founded by Meghnad Saha (*Resonance*, Vol.3, No.5, 1998) and Prafulla Chandra Ray (*Resonance*, Vol.6, No.1, 2001). It is clear from the articles adjoining this one in the same issue, that this journal had a wide scope. The fact that the authors did not go to a specialised physics journal reflects their conviction that the result was of wider significance. Also, that they chose an Indian – indeed Calcutta-based – journal reflects, to me, the spirit of those times, when there was a strong sense of building a new nation. The Russian group who published earlier, had in fact, sent cables (i.e., telegrams!) to *Nature*, which is again a broad journal covering all of science. Interestingly, C V Raman from Kolkata had used the cable strategy to establish priority over the Russian group in 1928. Although the paper is a brief communication, it has a clear introduction to the phenomenon of fission, which was only five years old at that time, with much of the work shrouded in wartime secrecy. It also has extensive follow-up analysis of which nuclei were produced in the fission process.

Reading this paper leaves us with two 'might-have-been' questions. First, if the authors had had the brash self confidence of a C V Raman, might they have been the first rather than the second to publish the lifetime of spontaneous fission? On the other hand, we must keep in mind that Raman knew that the phenomenon he was looking for was predicted by theory, while the lifetime measured in this work was far shorter than the simple theory. Second, was the later follow-up commensurate with such a fine beginning in experimental nuclear physics? The answers to both these must be sought in the domain of the history and sociology of science!

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Two sets of experiments were carried on, one with dry seeds and the other with seeds soaked in water for a period of 6 hours. Seeds were treated with different doses of X-rays varying from 202.5 r to 6720 r and three different kinds of rays were given, namely, soft, medium and penetrating in different cases.

A detailed account of the results obtained will be given in a fuller paper to be published elsewhere, but a summary of the results is given as follows. (1) Plants raised from irradiated seeds appear to lose their vitality during early stages of their growth but later on their growth rate becomes enhanced. (2) Some of the irradiated plants show bifurcation of the tip at an early stage of growth and some assume a bushy appearance on account of vigorous lateral branching. (3) Some plants have two or three leaves in one internode in contrast to the normal alternate phyllotaxy of the plant. (4) In some cases delay in flowering caused by irradiation is marked. (5) Amount of woody tissues (xylem) increases due to irradiation. (6) Number of fibre cells appear to be increased in some cases, but the shape and size of the fibres are found to be unchanged. (7) Action of X-rays on moist seeds is more vigorous than that on dry ones.

The present study on the effects of X-rays on Jute seeds both dry and wet show some characteristic changes though none of them is dominant and no particular effect is common to every individual. In our experiments the rate of germination was enhanced in the seeds of *Corchorus capsularis* treated with soft heavy doses and light penetrating doses, but no such effect was observed in those of *C. olitorius*. According to Altman⁷ and others, and Koernicke, growth of seedlings became rapid when they were treated with weak doses of X-rays; but I have found that the rate of growth of all the irradiated seedlings was very slow to start with but after a time they revived and began to grow vigorously. Johnson observed changes in the shape and size of the leaves; but no such change has been observed in jute plants.

I take this opportunity of thanking Dr N. Das Gupta of the University College of Science, Calcutta for kindly making arrangements for exposing the seeds to the action of X-rays. The exposures were given in the laboratories of the University College of Science, the Science Association and in the Chittaranjan Sevasadan.

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ON THE SPONTANEOUS FISSION OF URANIUM

DURING the course of their investigation on the chemical properties of the so-called "transuranic" elements, Hahn and Strassmann¹ suggested that uranium nucleus breaks up into two parts of comparable sizes, a barium nucleus ($Z=56$) and a krypton nucleus ($Z=36$). The first theoretical interpretation of this observation was given by Meitner and Frisch². According to them the nucleus is like a liquid drop subject to the following forces: (i) a short range non-electrical force, only acting between the neighbouring particles; this is proportional to the total number A of the particles in the nucleus, (ii) a force analogous to surface tension, also proportional to some power of A and (iii) a repulsive coulomb force, proportional to the square of the number of protons Z , and is opposed to the surface force. Just as a drop of liquid, when set into vibration may split into two drops, so might a nucleus. This becomes more probable for heavy nuclei, because of an effective reduction of surface tension, resulting from increasing nuclear charge. The actual nucleus will be stable so long as the sum total of the electrostatic and surface tension energy has a minimum for the spherical shape. With increasing size and charge of the nucleus, this minimum would flatten and would be expected to disappear for some critical value of Z . Nuclei of greater Z would break apart. Bohr and Wheeler³ have considered the stability of a nucleus (A, Z), against small arbitrary deformations, and have shown that there is a limiting value of $\frac{Z^2}{A}$, beyond which a nucleus is no longer stable with respect to deformations of the simplest type. They show that this ratio is greater by 17 per cent than the value of $\frac{Z^2}{A}$ for U^{238} . Such nuclei are therefore near the limit of stability and it is possible to calculate the potential energy necessary to deform the nucleus sufficiently to produce division. The value

of this deformation potential has been calculated for Th, Pa and U nuclei and they are found to vary between 6'0 (Th²³²) to 5'0 (U²³⁵) MeV. When a neutron is captured by such a nucleus, a compound nucleus in an excited state is formed and if the energy of excitation is greater than the energy of fission E_f , fission will usually occur. The detailed comparison of the calculated probabilities of different U, Pa and Th nuclei lead to good agreement with experimental results. The results obtained so far are based purely upon classical mechanics consideration which is in agreement with the fact that the zero point energy of the nucleus is found to be about 1/15th of the energy of fission E_f . The statistical distribution in size of the fragments of fission depends on the complicated dynamics of the dividing nucleus. The theory is not developed enough to give this distribution, but does indicate that there is a wide range of possible fragments even for energies slightly greater than the critical energy.

Turning to the quantum mechanical aspect of the problem, we have to consider the possibility of the "tunnel effect" which will make it possible for the nucleus to divide even in its ground state by passage through a portion of configuration space where classically the kinetic energy is negative. Bohr and Wheeler have calculated the fission probability λ_f on certain simplifying assumptions which include that the expression (V-E) in the Gamow function is of the order of E_f , i.e., 6 MeV. The mean life is found to be of the order of 10^{22} years. This calculated value is however only approximate.

The nature of the fission products induced by neutron bombardment has been investigated by either chemical or physical methods. In the former method small amounts of chemical elements are introduced as carriers and are then separated by the known analytical methods. The radioactivity of the products are studied with the help of G-M counters. This method gives only the chemical nature (dependent on nuclear charge Z) and not the mass A of the fission products; the latter must also be radio-active to be detectable. By this method, 80 different kinds of atoms belonging to 23 different elements have been detected.¹ The physical method depends upon the simultaneous determination of the ionisation produced by the two fission products. From the conservation of momentum between the fission products it follows that the kinetic energy of each is inversely proportional to its mass. This method gives the mass and not the nuclear charge of the fission products and can be used to detect both stable and unstable fission products. Jentschke² and Prankl³ have identified the following nuclear fragments produced by predominantly thermal neutrons:

Heavy—Pr, Nd, element 61, Sm, Eu, Gd in the order of decreasing abundance.

Light—Rn, Rh, Pd etc.

Spontaneous Fission Products.—Libby⁴ searched for radioactive iodine in a sample of pure uranium nitrate solution kept undisturbed for five years; he came to the conclusion that the spontaneous fission, if any, must have a half-life period of over 10^{14} years. Experimental evidence for the spontaneous fission of uranium nucleus was first reported by Petrzhak and Flerov⁵. They used as an ionisation chamber, a 15 plate condenser coated with thin layers of U₃O₈ and connected it to a high gain amplifier of extremely high resolution. They found evidence of such fission occurring at a frequency of about six per hour. By means of subsidiary experiments they concluded that the pulses could only be due to spontaneous fission, as their number was too large to be accounted for as being due to atmospheric neutrons. The half life of the U nucleus was calculated to be of the order of 10^{14} years.

One of us by using an essentially similar arrangement, the details of which will be published in the *Transactions of the Basic Research Institute*, has substantially verified the above findings viz. that spontaneous fission of U nucleus occur, which are not due to atmospheric neutrons and the average half life is $\approx 3 \times 10^{14}$ years. The ionisation pulses are counted either by a suitably biased thyatron relay connected with a telephone counter or are recorded photographically on a slowly moving cinefilm by a loop oscillograph. An arrangement is being set up for the direct measurement of the energy of the fission products.

If the above conclusion is correct, then in very old minerals rich in uranium, chemical analysis should indicate the presence of stable fission products. Recently, a small quantity of a highly concentrated uranium mineral containing material of orange colour (density 5.64, hardness 3) from some unknown locality in Rajputana came to our hands. It was found to be three times as radioactive as standard U₃O₈, and appears to be a variety of Gummite. The mineral was subjected to usual group separation and the groups to careful spectrographic analysis. The following table gives a partial analysis of the mineral:

UO ₂	H ₂ O	PbO	SiO ₂ &c.	ThO ₂
75%	7.25%	8.26%	2.23%	0.66%
Rare Earths	CaO	Na ₂ O	K ₂ O	Gases
0.38%	1.67%	0.70%	1.08%	0.20%
	Fe ₂ O ₃		Al ₂ O ₃	
	0.20%		0.20%	

The lead ratio is 0.1225. The corrected age appears to be about 847 million years. The radium content, calculated from uranium content is about 206 mgms. per ton.

Spectrographic analysis revealed the presence of Ge, Sb, Bi, Sr, Ag, Yt, Yb, Gd, La. Absorption spectra measurements (using the micro-technique of Hopkins), showed the presence of Nd and Pr. Ce was detected by spot-test. It is believed that some of the above elements are fission products.

A comparative chart of elements, formed by fission of uranium nucleus and identified by different methods is given here:

A. Induced fission:

(1) Chemical examination: Radioactive isotopes of

34Se , 85Br , 86Kr , 87Rb , 88Sr , 90Y , 40Zr , 42Mo , 47As ,
 59Ba , 61La , 52Ce , 57Pr , 54Xe , 55Cs , 56Ba , 57La .

(2) Physical measurements:

Lighter fragments with masses from 74 to 109, including

34Se , 44Br , 45Kr , 46Rb

Heavier fragments with masses from 127-162, including 59Pr , 60Nd , element 61, 62Sm , 63Eu , 64Gd .

B. Spontaneous fission:

(3) Chemical examination:

32Ge , 38Sr , 39Y , 50Rb , 51Cs , 57La , 58Ce ,
 59Pr , 60Nd , 64Gd , 70Yb .

It is well-known that radioactive minerals are seldom free from rare earths. It is also known that older uranium minerals are richer in rare earths than the young ones, though there is no apparent proportionality between the rare earth and lead content. It is suggested that the growth of some of them is due to the accumulation of fission fragments. The above statement can hardly however, be made very rigorously. For, very complete analyses of uraninites have been made by Hillebrand, Marsh, Hauser and others. Their results show that the abundance of the individual rare earth elements varies widely in minerals of different localities and sometimes occur in large amounts indicating that they form the original constituents of the mineral.

Our expectation that stable fission products should occur in old uraninite minerals can be tested quantitatively in the following way. The amounts of some of the so-called fission products in several such minerals are to be determined chemically. If it is found that in some of the minerals, two or more of the so-called fission elements occur in approximately equivalent weights, then the occurrence can be taken as an indication that they are produced by fission of the uranium nucleus of the same main life. For such determinations large quantities of uraninite minerals are required; these are at present not available.

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PHOSPHORESCENCE AFTER GLOW OF SOME ALKALI HALIDES

CRYSTALS of sodium chloride, potassium chloride and potassium bromide are fluorescent under X-ray irradiation. If the period of exposure be sufficiently long these crystals are also discoloured. The discoloured crystals possess extra absorption bands (F-centres). The same centres can also be produced by keeping the crystals in the vapour of the alkali metals at a high temperature. The mechanism suggested for this colouration is as follows: The ionic crystals contain usually a number of vacant lattice points (positive and negative ionic positions) and a number of interstitial atoms frozen in the lattice (in the way of Schottky and Frenkel defects). The photo-electrons released by X-rays execute a sort of Brownian motion before they get trapped in one of the vacant lattice points, formerly occupied by a negative ion. These trapped electrons can be raised to the conduction band by absorption of a suitable radiation and give rise to a new absorption band. To account for the emission we can assume that electrons are primarily raised to the conduction band and thus carried to the neighbourhood of the interstitial ions where additional level systems are present; next transition of electrons to lower levels may occur with the possibility of emission.

The electrons raised to the conduction band in their passage to the radiating centre may get trapped in trapping centres. In ionic crystals the following trapping centres are theoretically possible:—

1. Vacant lattice points (generally negative ionic positions),
2. Tamm's surface levels,
3. Landau's trapping centres.

The centres of the type (1) give rise to the F and F' centres; for the existence of (2) and (3) decisive experimental evidence is not yet available.

It has been observed that the after glow period for crystals of these compounds can be increased to