# Isenthalpic Flow, Joule–Kelvin Coefficients and Mantle Convection

WALDBAUM[1] has suggested that, when mantle material convects according to the model of Turcotte and Oxburgh[2], the temperature of an isolated rising mass will increase, "even in the idealized case where friction, viscosity and turbulence are ignored". He argues from the premise that the steady adiabatic flow of a fluid is isenthalpic along streamlines: the Joule–Kelvin coefficient of the fluid can therefore be used to predict the temperature change during decompression.

This argument is incorrect. Waldbaum has apparently been misled by variations in the published definitions of enthalpy. Although the enthalpy of a substance is always defined by an expression of the form

$$h = u + pv \qquad (1)$$

($h$ = specific enthalpy, $u$ = specific internal energy, $p$ = pressure and $v$ = specific volume), there are important variations in the definition of the internal energy $u$. Of the two authorities quoted by Waldbaum, Pippard[3] includes in $h$ and $u$ terms for kinetic and potential (gravitational) energy, whereas Liepmann and Roshko[4] include a term for kinetic energy in what they call "total enthalpy", but make no mention of potential energy. For them and for Landau and Lifshitz[5], however, the unqualified term enthalpy is taken to describe an intrinsic property of a substance, dependent only on local thermodynamic variables; it is "the enthalpy which would be measured by an 'observer' moving with the fluid"[4]. Pippard's definition permits him legitimately to say that, in steady adiabatic flow, the enthalpy of a fluid is constant along streamlines. For Landau and Lifshitz, however, the quantity which is constant is the sum

$$\text{enthalpy} + \text{potential energy} + \text{kinetic energy} \qquad (2)$$

It can now be seen why the Joule–Kelvin coefficient $(\partial T/\partial p)_h$ is not directly applicable to free convection in a gravitational field. This coefficient is defined physically for a condition of constant intrinsic enthalpy, which does not exist where decompression occurs through upward movement in a gravitational field. For a slowly moving frictionless fluid, the stress field in those circumstances must be purely hydrostatic, and in adiabatic flow the entropy ($s$) will be constant. Differentiating (1) and (2) gives

$$\mathrm{d}u + p\,\mathrm{d}v + v\,\mathrm{d}p + g\,\mathrm{d}z = 0 \qquad (3)$$

where $g$ is gravitational acceleration and $z$ is vertical height. As $v\mathrm{d}p = -g\mathrm{d}z$ in hydrostatic conditions, we obtain the standard equation of energy balance for reversible adiabatic expansion

$$(\mathrm{d}u + p\,\mathrm{d}v)_s = 0 \qquad (4)$$

The temperature gradient is therefore the well known adiabatic gradient of geophysical fluid dynamics, based on the isentropic coefficient $(\partial T/\partial p)_s$.

It might be suggested that isenthalpic heating or cooling coefficients could be applied to deviations of the stress field in a real fluid from a vertical hydrostatic gradient. Such deviations could be considered to correspond to the irreversible "isenthalpic steps" of Walbaum's argument. But how large are they ? The topography of the mid-ocean rift suggests ~0.6 kbar near the surface, whereas Mackenzie[6] obtained values less than 1 kbar at depths of 50 to 100 km under trenches from an analysis of gravity anomalies. Waldbaum's isenthalpic coefficients therefore suggest temperature rises of ~20 K or less. It is also questionable whether the flow is truly adiabatic in the regions of maximum non-hydrostatic stress; as Tozer[7] points out, when viscosity of a fluid varies rapidly with temperature, shear flow and associated viscous dissipation will tend to be strongly localized. The evidence of high heat flow behind island arcs tends to support this view[8]. Thus it seems likely that Joule–Kelvin coefficients will be of limited value in estimating temperature changes related to slow planetary convection.

Because of these problems it seemed worthwhile to examine further the variations in usage of the concept of internal energy among thermodynamicists. Many workers bypass the problem by not mentioning gravitational and kinetic energy at all. Gibbs[9] clearly separated gravitational energy from what he called the "intrinsic potential" of a chemical component of a thermodynamic system: he included kinetic energy[10], only to show that it must vanish at thermodynamic equilibrium. Guggenheim[11] treats gravitational energy in the same way as Gibbs, but Lewis and Randall[12] take the alternative approach and include it in the internal energy. It is unfortunate that few authors except Lewis and Randall[12] and Denbigh[13] point out the existing divergences, for there is evidently little prospect of reaching a consensus on this matter in the context of chemical thermodynamics. Considering the thermodynamics of fluid flow (a subject not usually discussed by chemical thermodynamicists) there is much to be said, however, for general adoption of the Landau–Lifshitz approach, because the kinetic and gravitational components of the total energy may often be analysed more or less independently of local thermodynamic parameters. In the absence of uniformity, confusion may be minimized by use of the adjectives "intrinsic" or "total" to indicate the sense in which the terms energy, enthalpy or chemical potential are being used in a particular context.

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# Spontaneous Fission Previously Observed in a Mercury Source

PREVIOUSLY[1], we have presented evidence for the possible existence of a superheavy element with atomic number 112. The principal evidence for the possible existence of this element was based on the observation of spontaneous fission in a mercury source separated from a tungsten target which had been irradiated by 24 GeV protons. Spontaneous fission activity was observed, using polycarbonate foils, in mercury sources separated from two tungsten targets. These latter are identified by the symbols W2 and W3. In the case of the W3 source the proton dose was probably less than for W2 and also only about 30% of the mercury added as carrier was recovered in the final source (as measured by colorimetric techniques). The observed fission activity was smaller than that observed with the W2 source. In the latter case (W2) the measured activity was originally about three fissions/day and all experimental work reported here has been carried out with this sample.

The argument that the observed activity might be due to a superheavy element was based entirely on the prediction that element 112 will be the chemical homologue of mercury.

Such an identification may, of course, be in error because of the possibility of contamination of the sources. This topic was considered in some detail in our earlier work. In particular it was shown that the most likely spontaneously fissioning contaminant, $^{252}$Cf (2.5 yr), could probably be excluded as there was no evidence for an associated alpha group (branching ratio 97%) at 6.1 MeV. Such a group would be expected to be sixteen times more intense than the observed rate of fission. In the case of the W3 target, immediately after the original chemical separation the Hg fraction gave $2.5 \pm 0.6$ fissions per week and the combined actinide–rare earth fraction gave $7 \pm 2.5$ fissions per week. Subsequent counting of X-rays in the Hg source indicated that less than 0.1% of all rare earths and actinides were present in the Hg fraction.

Since the earlier experiments, attempts have been made to identify the source of the observed fission activity more directly by trying to measure nuclear properties which depend in some way on the $Z$ or $A$ of the fissioning nucleus. As will be shown, this attempt has only been partially successful and the present letter is in the nature of a progress report on some continuing experiments, the results of which are of general interest.

With the aim of being able to measure the energy spectrum of the fission fragments some attempts were made to reduce the thickness of the W2 source and to make it more uniform. For these reasons the source was ozonized and mechanically spread on the Pt backing plate. While the source was still too thick to permit accurate measurement of the fission fragment energy spectrum, it was possible both to detect the fission fragments and measure the alpha particle energy spectrum simultaneously in the same silicon surface barrier detector. With this system we measured an upper limit of 5 : 1 for the ratio $\alpha/f$, of all alpha particles having energies near 6.1 MeV to spontaneous fission activity. This indicated that at that time at most 30% of the spontaneous fission activity could have been due to $^{252}$Cf. The number of fissions in the source was frequently measured with polycarbonate films over a period of 3 months after ozonization and gave an average value of $4 \pm 0.3$ fissions per day over this whole period. Statistical uncertainties made it impossible to determine whether the fission activity was growing or decaying.

After this period a more uniform source was prepared by chemically exchanging the Hg activity onto an evaporated film of PbS on a $240 \pm 20$ µg cm$^{-2}$ polycarbonate backing. Roughly 50% of the fission activity was transferred onto the PbS film. With this source it was now possible to measure the kinetic energy spectrum of the fission fragments and energy spectra were measured with two silicon surface barrier detectors for both single and coincident summed kinetic energies. The observed spectra are shown in Fig. 1. We also show in Fig. 1 the spectra obtained in the same apparatus from a $^{252}$Cf source which was prepared by sublimation of $^{252}$Cf onto successive layers of PbS to simulate the HgS–PbS source as closely as possible. Clearly source thickness and non-uniformity could greatly affect the spectral shapes.

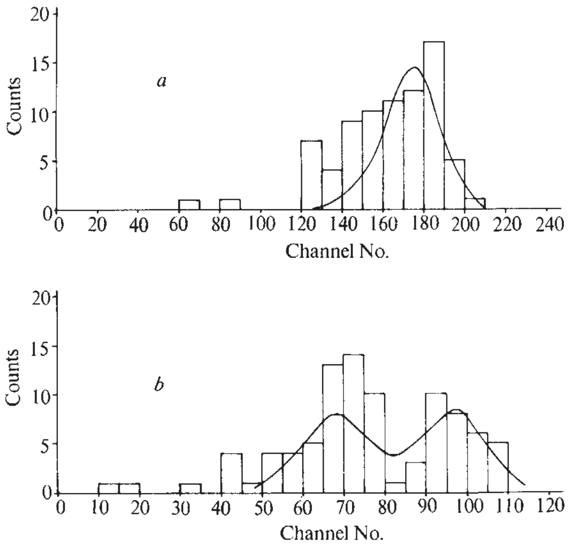

**Fig. 1** Energy spectra of fission fragments observed with the mercury source. In *a* the summed kinetic energy spectrum is given, and in *b* the singles spectrum observed with the front detector. The continuous lines show similar measurements with a $^{252}$Cf source with much better statistics.

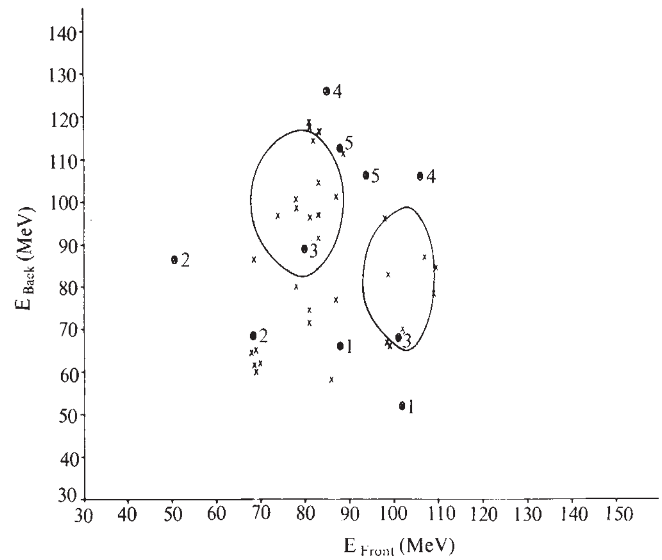

**Fig. 2** Correlated energies of fission fragments from the Hg source. The pairs of numbered points show the alternative interpretations when the analysis of the events is ambiguous. The contours enclose the region in which 90% of $^{252}$Cf fragments would be found and are asymmetric because of energy loss in the source backing.

Fig. 1*a* shows that the two sum spectra appear to differ in shape particularly in the low energy region and that the Hg sum energy spectrum seems to have a peak at slightly higher energy than that for $^{252}$Cf (roughly 194 MeV, as compared with 182 MeV for $^{252}$Cf). Because of the difference in spectral shapes, however, the average value of the sum kinetic energy for the Hg source is lower than that of $^{252}$Cf. The possibility of non-binary fission cannot be ruled out for superheavy elements, so any conclusions based on the assumption of binary fission must be regarded with caution. In addition, if multiple fission occurs, our two detector system will not in general measure the total energy. With this in mind, it is worthwhile noting that since the total kinetic energies for binary fission are roughly proportional to $Z^2/A^{\frac{1}{3}}$, then fragments from the binary fission of element 112 would be expected to have a total kinetic energy 24% higher than that for $^{252}$Cf (or about 225 MeV) whereas those from Hg isotopes would be expected to have a total kinetic energy of about 130 MeV. A statistical $\chi^2$ analysis indicates that the probability that the observed sum spectrum is consistent with that of pure $^{252}$Cf is less than 1%. This probability increases to 30% if the point with the largest value of $\chi^2$ (at channels 120–130) is removed from consideration.

The energy spectrum of single fission fragments from the W2 source is shown in Fig. 1*b*. The two peaks in the spectra are in similar positions to those measured for a $^{252}$Cf source under the same conditions. The ratio of the heights of the high energy to low energy peaks is $0.7 \pm 0.2 : 1$ for the Hg source compared with a value of 1 : 1 measured for the $^{252}$Cf source.

The fission data were printed out sufficiently often that about 50% of the individual singles and coincidence events could be correlated with each other. In these cases, it is possible to determine the energy of the fission fragments striking the back detector by a simple subtraction. The energies of the correlated fragments are plotted in Fig. 2, together with a contour

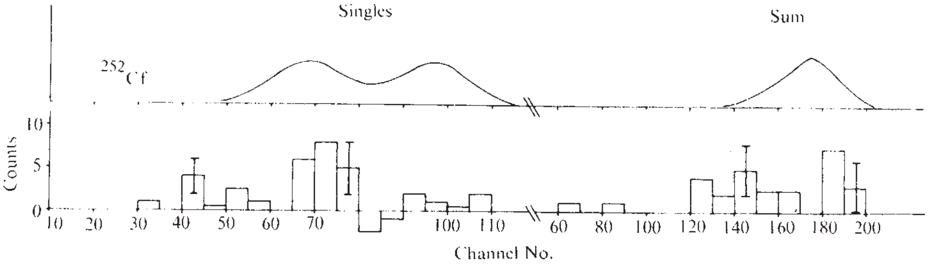

**Fig. 3** Fission energy spectra for coincident and singles events after subtraction of a $^{252}$Cf kinetic energy spectrum normalized to 70% of the total number of fission counts. Also shown for comparison are spectra obtained with a $^{252}$Cf source.

which encloses the region in which 90% of the $^{252}$Cf fragments would be found. The relatively large number (30%) of the observed fissions from the Hg W2 source which do not fall within the contours can be regarded as evidence of spontaneously fissioning material other than $^{252}$Cf in the Hg W2 source.

Using the same thin mercury source another measurement was made of the ratio of intensity of 6.1 MeV alpha particles to fission fragments. The value obtained was $\alpha/f = 11 \pm 1.5$ to be compared with the known value $\alpha/f = 16$ for $^{252}$Cf. If the 6.1 MeV alpha particles are associated entirely with the disintegration of $^{252}$Cf this would imply that ~70% of the observed fission activity at that time stemmed from $^{252}$Cf.

Using the same W2 mercury source a measurement of the mean number of neutrons emitted per fission ($\bar{\nu}$) was made using the apparatus described by Mather *et al.*[2]. In the present measurements the requirement for a coincidence between the fission fragment and a prompt gamma ray was omitted. The value of $\bar{\nu}$ obtained in this way was $3.7 \pm 0.5$ which when compared with the known value of $\bar{\nu} = 3.76$ for $^{252}$Cf again suggests that a significant fraction of the activity could have been due to this element.

Measurements of the fission energy spectra, the mean number of neutrons per fission and the alpha/fission intensity ratio all suggested that a significant fraction of the observed fission activity from the thin source could have been due to $^{252}$Cf. Further chemical separations were therefore carried out on the Hg source from the W2 target to give both mercury and actinide fractions. Measurements of the fission and alpha activity of the resulting actinide source showed that $73 \pm 7$% of the fission activity observed at the time of the kinetic energy measurements now appeared in the actinide fraction and here the 6.1 MeV alpha to fission ratio was $16 \pm 2 : 1$ indicating that this fission activity was probably due to $^{252}$Cf.

With the Hg fraction an attempt was made to exchange the Hg onto a PbS coated film. Approximately 70% of the original Hg tracer and no detectable (< 1%) rare earth tracer were found in this HgS–PbS source. Using a surface barrier detector in the same geometry as for the original source about 3% of the fission activity ($1 \pm 0.5$ per week) was found in the HgS–PbS source. The ratio, $\alpha/f$, for this source for the few alpha particles observed between 6.05 and 6.15 MeV was found to be $4.7 \pm 4 : 1$. A subsequent 2 week measurement of the fission activity of this source with a polycarbonate film gave no events. The source from the residual solution was too thick to be counted.

From these measurements it appears that approximately 70% of the fission activity now observed from the W2 source is due to $^{252}$Cf although the remaining 30% seems unlikely to be due to this element. As early alpha spectra showed smaller amounts of alpha activity having an energy of around 6.1 MeV for comparable rates of fission activity we conclude that after the original separation either the source has been contaminated or the $^{252}$Cf has grown in. The frequent changes in source geometry and low counting rate make it difficult to distinguish between these possibilities. It is not possible to explain the changes in alpha to fission ratio as being the result of changes in source thickness as the alpha particles have a range which is larger than that of the fission fragments.

Finally, in Fig. 3 we show the results of the fission energy spectra measurements after subtraction of a $^{252}$Cf kinetic energy spectrum normalized to 70% of the total number of fission counts. Within the very large statistical uncertainties of these residual spectra it appears that the total kinetic energy spectrum either shows two peaks (one broad peak centred about 150 MeV and one at about 195 MeV) or one very broad peak (centred at about 170 MeV). The single spectrum shows three regions: two broad regions centred at about 65 and 100 MeV and a sharper and more pronounced peak at 85 MeV.

The Hg W2 source used for the kinetic energy measurements was placed on an Ilford K-1 nuclear emulsion for 13 days before the final chemical separation. Examination of the tracks in this emulsion gave an upper limit of 0.5 non-binary fissions per day (about 30% of the "non-Cf" fissions).

The fact that the measurement of $\bar{\nu}$ for the total Hg source was close to that of $^{252}$Cf indicates that the remaining 30% of the fission activity has a neutron multiplicity in the range of 2 to 5. The distribution of multiplicities did not indicate any events of multiplicity 7 or greater and also no large preponderance of events of multiplicity 1.

To summarize, both the alpha to fission ratios and subsequent chemical separations show that at the time of our kinetic energy measurements 70% of the fission activity was due to $^{252}$Cf. After subtraction of the $^{252}$Cf component the residual fission kinetic energy spectra differ considerably from those of known actinides. An unlikely explanation is that the distribution of kinetic energies could indicate a mixture of spontaneously fissioning species, one in the light Hg-Po-Ra region (but see ref. 3) and one in the heavier No-Lw region. Another possibility is that it could be the spectrum of fragment energies produced in an unusual mode of fission such as predicted by Greiner[4] for superheavy elements. It must be noted that the kinetic energy spectra and the value of neutron multiplicity do not agree with the published predictions for binary fission of superheavy elements; however, neither do the kinetic energy spectra appear to fit those of any known spontaneously fissioning actinide isotope.

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# Complete Fusion induced by Krypton Ions: Indications for Synthesis of Superheavy Nuclei

COMPLETE nuclear fusion has been produced by bombarding targets of germanium, cadmium and thorium with 450 MeV $Kr^{23+}$ and 500 MeV $Kr^{24+}$ obtained from the Orsay accelerator ALICE[1]. Recoiling synthesized nuclei were slowed down in helium gas, transferred through a capillary tube and deposited on a steel rod in an evacuated enclosure[2]. An annular silicon semiconductor counter around the capillary tube detected the alpha particles produced by the decaying nuclei. Typical bombardment times were of the order of 10 h with $10^8$ Kr ions $s^{-1}$, and the background was shown to be insignificant by bombarding for 24 h without a target in place. Further tests also showed that secondary neutrons and gamma rays did not induce nuclear reactions in the detector.

The apparatus was tested by carrying out experiments with an argon beam and a dysprosium target[3] that produced the known alpha emitters $^{200}Po$ and $^{199}Po$. Also the reaction $^{84}Kr + ^{74}Ge \rightarrow ^{153}Er + 5n$ was studied and found to give a maximum cross section of 20 mbarn at a bombarding energy of 370 MeV. The total cross section for $^{74}Ge$ (Kr, $x$n) reactions of 32 mbarn was found to be in agreement with theoretical estimates based on the concept of critical angular momentum[4,5]. This shows that the total fusion can be represented by:

$$\sigma_{CF} = \pi\hbar^2 \frac{l^2_c}{2\mu\bar{\varepsilon}}$$

where $l_c$ is the critical value of the angular momentum quantum number, $\mu$ is the reduced mass and $\bar{\varepsilon}$ the centre of mass energy. Studies of the reactions $^{116}Cd + ^{84}Kr \rightarrow ^{197}Po + 3n$ at a bombarding energy of 375 MeV and $^{164}Dy + ^{40}Ar \rightarrow ^{200}Po + 4n$ at 190 MeV gave cross sections of 3 mbarn and 25 mbarn, respectively, demonstrating the effect of the enhancement of the fission width because of the larger angular momentum of the krypton projectiles, and the decrease of fission barriers for light polonium isotopes.

After the successful demonstration of the technique and with the confidence placed in the theoretical prediction of the cross sections by the above results we bombarded a thorium target with 450 MeV $Kr^{23+}$ ions for 10 h. No activity was observed—probably because the energy was near the Coulomb barrier—and the experiment was repeated with 500 MeV $Kr^{24+}$ ions on a 1 mg $cm^{-2}$ thorium target. The alpha spectrum obtained at this energy is shown in Fig. 1. Energy calibrations with the thoron deposit (8.78 and 6.05 MeV) and with $^{153}Er$ (4.67 MeV) and $^{152}Er$ (4.80 MeV) are shown with larger arrows. Many alpha particles of energy between 7 and 9 MeV have been assigned to light isotopes of polonium, astatine or radon which might have been produced by fission of very heavy nuclei. The absence of $^{212}Po$ at 8.78 MeV is significant. The peaks at 8.1 MeV, 8.48 MeV might be attri

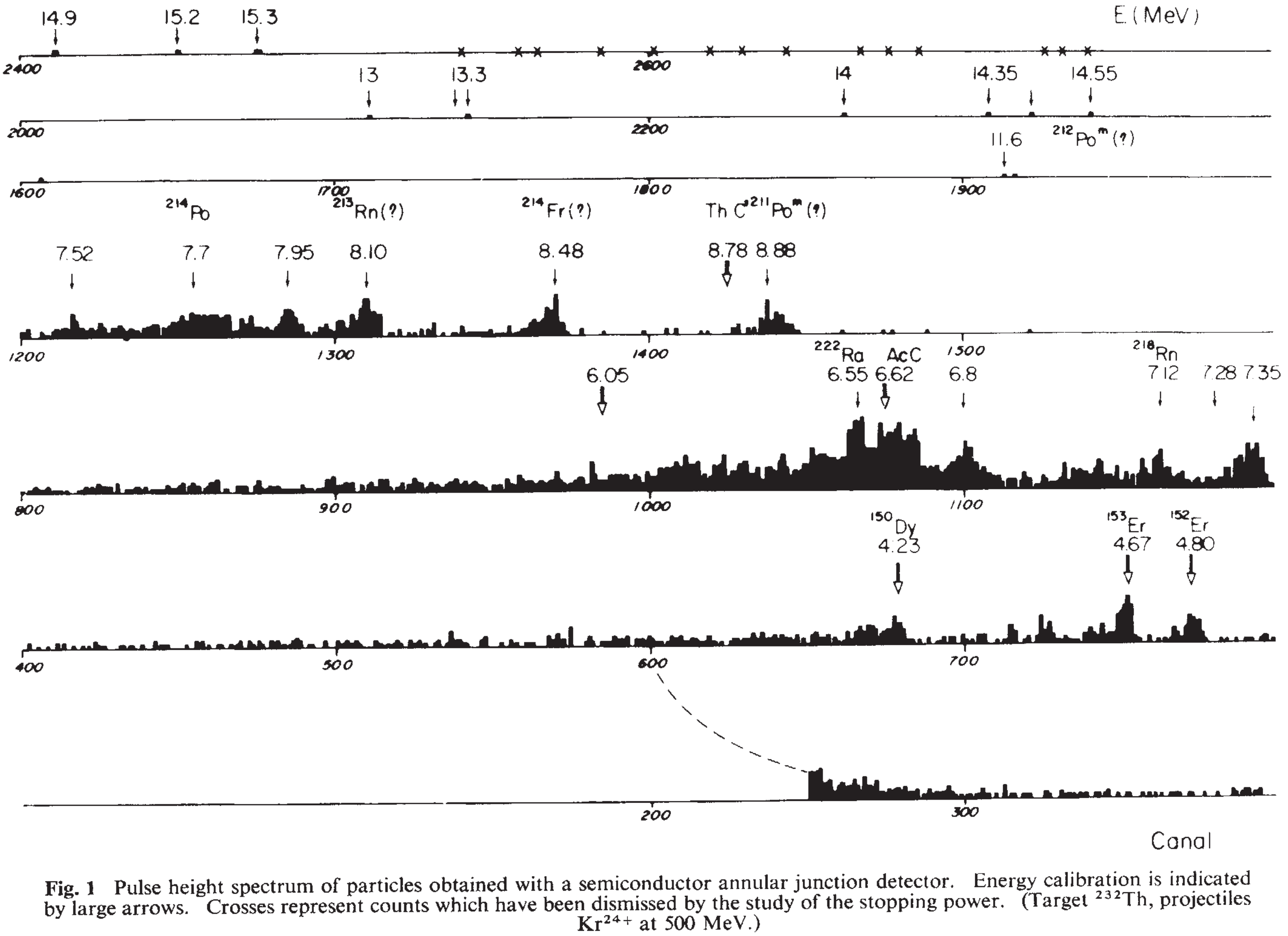

**Fig. 1** Pulse height spectrum of particles obtained with a semiconductor annular junction detector. Energy calibration is indicated by large arrows. Crosses represent counts which have been dismissed by the study of the stopping power. (Target $^{232}Th$, projectiles $Kr^{24+}$ at 500 MeV.)