Measurements of fission yields

H. O. Denschlag

Institut für Kernchemie, Universität Mainz, D-55128 Mainz, Germany

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Introduction

The discovery of nuclear fission was certainly one of the most important findings of the 20th century. It had a strong impact on the economic and military situation of the world in the second half of the century. Beside these practical consequences, the detailed study of the fission process has contributed very fundamental insights into the structure of matter, in particular on the interplay of liquid drop- and shell- (particle- and wave-) properties.

The sensational discovery made on December 17, 1938 by O. HAHN and F. STRASSMANN\textsuperscript{1,2} was based on the chemical identification of barium isotopes as products formed in the irradiation of uranium by neutrons.

Early measurements

The first semiquantitative (mass) yield measurement was already published two months later in February 1939 by W. JENTSCHKE and F. PRANKL.\textsuperscript{3} It was carried out using an ionisation chamber detecting the energy of the fission fragments. It showed the asymmetric nature of the fission process with a mass ratio of 1.60\pm0.06, corresponding to two maxima of the mass yield curve around $A = 139$ and 87. The mass difference between the compound nucleus and both fragments was interpreted as a hint for neutron emission in the fission process. Several attempts\textsuperscript{4,5} to measure directly the number of neutrons set free in the fission process led to first approximate numbers.

Such an emission of neutrons in the fission process was highly interesting because it could allow to realise a self sustaining chain reaction. FLUGGE\textsuperscript{5} reports already in June 1939 that an amount of 4 tons of uranium submitted to fission could produce the German requirement of electricity (at the time) for a duration of 11 years – the author adds the somewhat amusing remark that, of course, once every day the engine has to be cleaned of fission products... On the other hand, he calculates that under certain conditions the total amount of uranium will have undergone fission within a time span of $10^{-4}$ seconds – "so that we have to do with an extremely strong explosion".

These insights that came to many scientists at the same time, led, in particular the US Government, to establish large radiochemical laboratories which were dedicated, in the frame of the secret "Manhattan Project", among other problems, to the measurement of fission yields.

The first radiochemical measurements with the goal to quantitatively determine fission yields were probably carried out by ANDERSON, FERMI, and V. GROSSE\textsuperscript{7} under adventurous conditions of acid fumes and other explosions.\textsuperscript{8} These measurements provided mostly the yields of the end members of $\beta$-decay chains that are formed directly in the fission process and by the decay of $\beta$-unstable precursors (chain yields or "cumulative yields" of the last chain members). They have been published\textsuperscript{9} in comprehensive form several years later.

The extension of these radiochemical yield measurements to short-lived isotopes at the beginning of the $\beta$-decay chains and a differentiation between the direct formation in the fission process ("primary" or "independent" yields) and a formation by the $\beta$-decay of precursors ("secondary" yields or "cumulative" yields [when adding the independent yields]), came, in general, only later with the development of fast (frequently automated) chemical separations\textsuperscript{10-13} and their application to the determination of fission yields.\textsuperscript{14}

Present status

Meanwhile the total body of available fission yield data has reached a number of about 60 000 values, that are compiled in extensive data banks\textsuperscript{15,16} covering about 60 different fission reactions (different fissile isotopes from thorium to fermium induced by different particles of varying energy etc.). A large fraction of these data have been obtained by purely physical methods, as the radiochemical techniques dominating in the early days have meanwhile been supplemented by physical techniques.
Lately, physical techniques have been developed that are superior to the radiochemical techniques with regard to the efficiency (number of yields determined in a certain time and by a certain manpower), to the precision of the values obtained, and to the half-lives of fission fragments accessible to such measurements. In addition, these physical measurements provide parameters that are not available in radiochemical measurements. For instance, fission yields can be measured as a function of the kinetic energy of the fission fragments, hence, providing information on the influence of the shape of the compound nucleus at scission. (A compact shape will lead to high kinetic energy fragments and a deformed shape will lead to low kinetic energy and correspondingly high internal excitation energy and a high number of prompt neutrons emitted).

The methods mentioned to measure fission yields and the results obtained have been reviewed several times recently. The topic of the present article will be – after the historical reminiscences made above and induced by the topic of this issue at the turn of the millennium – to present a few very recent results and/or novel techniques and, with these examples in mind, to give an outlook for problems to be faced and techniques to be used in future.

Recent trends and outlook

Motivation

It may be surprising that in spite of the wealth of more than 60,000 yield values available, there is still a need to do more experiments and to measure additional values. This need and the interest exists like in the early days of the pioneers and it is motivated by the same two reasons: fundamental interest in the structure of matter and practical applications of nuclear fission.

The interest motivated by practical application in our days are novel alternative reactor types, e.g. accelerator driven reactors, possibly based on the breeding of thorium and using a melt of fluorides, with an online separation of fission products (the “daily clean-up” of the engine as proposed by Flügge in 1939). The neutrons in such a system are produced from 1 GeV-protons impinging on a lead target or directly on some fissile material, hence, a new interest in high energy spallation and fission reactions and yields. Concepts relying on thermal and fast neutron fission are being discussed. Dedicated systems for the transmutation of “minor actinides” for safety reasons require exact information on delayed neutron emission and on decay heat and, therefore, a knowledge of the yields of short-lived fission products.

The interest motivated by fundamental questions is probably best illustrated by some recent experimental results and instrumental developments.

Lohengrin

The first example concerns pairing and shell effects in the fission process. The asymmetric character of fission discovered very early was a mystery until the role of the double shell closure at $A = 132$ ($Z = 50$, $N = 82$) was discovered: The tendency of the fissioning nucleus to preserve intact a core of this mass leads to an asymmetric scission configuration and to an asymmetric yield distribution. Ever since, one was asking the question whether there would be an influence of the other double shell closure in the mass range of the fission products at $^{78}$Ni ($Z = 28$, $N = 50$). The very low background of the mass separator for unsloshed fragments Lohengrin allows one to descend to very low yields and correspondingly to low mass numbers of the fission products. Mass yields measured recently using Lohengrin in this low mass region are shown in Fig. 1.

In all the measured fission reactions shown, a hump is observed at mass 70. The isotopic composition of the hump has been measured using a technique based on the specific energy loss of fission fragments in matter. It shows almost exclusively (95% of the chain yield) $Z = 28$ (Nickel) ! In consequence, this seems to be the sought effect for the shell closure at 28 protons. The effect appears to be less pronounced for $^{240}$Cf. This is in agreement with the expectations for typical shell effects, that are known to diminish for heavier fissile systems because their higher fissility leads to an increased excitation energy at scission.

What about the shell closure at $N = 50$? The isotope $^{70}$Ni has 42 neutrons and is 8 neutrons away from this closed shell.

Looking more closely at Fig. 1, a weak kink is observed at mass 80 at least for the isotopes $^{233}$U, $^{239}$Pu, and $^{242}$Am. In this less asymmetric region around mass 80 more than the four fission reactions displayed in Fig. 1 have been measured. Therefore, Fig. 2 shows this mass region again with the complete experimental information. In addition, in this figure the data points above mass 79 have been fitted to a straight line (in this logarithmic display) in order to show more clearly the breaking away of yield for masses below $A = 80$. Considering the compressed logarithmic scale of the yield axis, the effect is quite large.