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CLUSTER RADIOACTIVITY: AN OVERVIEW AFTER TWENTY YEARS

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Abstract. The present status of experimental research in cluster radioactivity is reviewed with emphasis on results obtained in the last few years. Various theoretical approaches are briefly discussed and compared with recently obtained experimental results on ^{34}Si and ^{22}Ne clusters. Experiments in progress on ^{238}U and ^{223}Ac are described, and open problems in the experimental and theoretical research are outlined.

Key words: cluster radioactivity, fission models, alpha decay models.

1. **INTRODUCTION**

Writing this paper brings us back to very exciting years when our experimental research on cluster radioactivity first started (1988) and subsequently grew up reaching a steady state in the period 1990–2000. What was peculiar of that period was first of all the novelty of the phenomenon at that time (1984) recently discovered and the consequent excitement which led several experimental groups at Berkeley, Dubna, Orsay and finally Milano to develop experimental techniques dedicated to investigate such peculiar, extremely rare decay mode of heavy nuclei. However, this was not the only motivation of an intense experimental investigation which, in the course of about twenty years allowed to measure the decay properties of some twenty trans-lead exotic emitters of clusters with mass numbers in the range 14-34. Indeed, most important was the close connection between experimental and theoretical groups, the latter providing the former ones with predictions, in the form of extensive tables or computer programs, which were extremely important in guiding experimental investigations among the thousands of cases corresponding to different combinations of mother nuclei-emitted clusters exhibiting a positive Q-value for such radioactive decay.

In this connection the theoretical work of Dorin Poenaru on cluster radioactivity stimulated, encouraged and addressed we experimentalists in the difficult job of finding out a needle in a haystack: it is well known indeed that the branching ratios relative to α-decay are vanishingly small, ranging between 10^{-9} and 10^{-16} for the cases measured up to now. Moreover, the branching ratio is not the only parameter to characterize the difficulty of this research: one should mention, among others, the problem of preparing a high enough number of mother nuclei in order to compensate for the corresponding small value of the partial decay probability, which has recently been measured down to 10^{-30} s^{-1}. It is indeed clear that the situation is completely different if the mother nucleus is member of a natural radioactive series (as it was true in the pioneering work of Rose and Jones [1], Price [2] and Tretyakova [3]), or has to be synthesized by means of artificial radioactivity or radioactive ion beam techniques, as was true for the most recently measured decays and will be shown in next section. Considering this and other difficulties involved, one can understand why the selection of the most promising cases based on a reliable theory is of paramount importance. Several review papers have been written in the past covering both theoretical and experimental aspects of cluster (exotic) radioactivity [4]. In this paper, we will limit ourselves to describing the most notable achievements in the experimental research which took place after 1999, when the most recent review was published [5].

2. EXPERIMENTS ON CLUSTER RADIOACTIVITY AFTER 1999

2.1. ^{34}Si DECAY OF ^{242}Cm

We discuss this experiment first [6] since it was crucial in discriminating between different theoretical descriptions of cluster radioactivity. It is well know indeed that cluster radioactivity occupies an intermediate position between alpha-particle radioactivity on one side and spontaneous fission on the other. These two extreme processes of hadronic decay of nuclei are usually described by completely different formalisms. The first one ("α decay"-like) is considered to be non-adiabatic [7, 8]. Its probability is determined by the overlap of the parent nucleus wave function with those of both fragments resulting in a sudden formation of a cluster which then attempts to penetrate the Coulomb barrier. The other one ("fission"-like), on the contrary, is thought to be adiabatic [9, 10]. It includes the prescission phase where the fragments are overlapping. It was indeed shown by D. Poenaru [11] that the transmission probability through that part of the potential barrier before the saddle point, i.e. the prescission phase, simulates the so-called spectroscopic factor present in the "α decay"-like models, implicitly assumed to be unity in the "fission"-like models.

Previous experimental studies on cluster radioactivity have allowed us to accumulate data on clusters with mass number in the range A=14-32 [5]. When compared with theoretical models it is found however that despite their seemingly different nature almost all of them reproduce the experimental half-lives with the same kind of accuracy, i.e. within one-two orders of magnitude, thus making practically impossible to decide in favour of one or another. However, from the experimental point of view the gap between alpha-radioactivity and fission remains far from being filled, and observation of heavier and heavier clusters is important in this respect. For example, calculations show that the decay probability decreases with increasing fragment mass more steeply for the α-decay rather than for the fission-like models [11].

Unfortunately in going to heavier mother nuclei not only has the experimentalist to face with vanishing decay probabilities but also and especially with increasing competition coming from fission fragments in addition to the well known one from α particles. This fact has considerably slowed down the experimental efforts in this field in recent years.

The aim of this experiment was to measure the cluster decay of ^{242}Cm, the heaviest mother nucleus studied in this respect. All the models predict that the most probable clusters should be ^{34}Si and ^{208}Pb respectively, a channel which maximizes the decay Q-value due to strong shell effects.

In this study a particular care was directed towards the fission fragment suppression, which is one of the major problems met by experiments on transuranic nuclei, to the level of $\simeq 10^{-9}$ relative to the initial one. This was achieved by filtering the fission fragments with different kind of absorbers, both solid and gaseous. As in previous studies, solid state track detectors of the glass type have been used due to their selectivity in respect to the less ionizing but much more abundant α particles. Particular care was also devoted to the track detectors calibration and to the control of the α fluence, which is a critical point when dealing with branching ratios relative to α decay ($B_\alpha = \lambda_{cl}/\lambda_\alpha$) values of the level predicted for the present experiment, $10^{-16} - 10^{-17}$.

The sources were prepared by irradiating a ^{241}Am sample with neutrons produced by the Kurchatov research reactor and separating Cm by means of radiochemistry techniques. They were used to irradiate two ensembles of track detectors arranged in hemispheric geometry. Subsequent chemical etching, detector scanning and comparison of the events found in the form of conic tracks with accelerator calibrations allowed to find and to attribute to Si clusters 15 events. From the known number of ^{242}Cm atoms B_α came out to be 1.0×10^{-16} and the partial half life $(1.4^{+0.5}_{-0.3}) \times 10^{23}$ s.

When comparing such data with predictions given by the various theoretical models, we see that while the one of the fission-like model of Poenaru [9] gives a remarkable agreement, within a factor of two, those of the "α decay"-like models [7, 8] exhibit the largest discrepancy, up to one-two orders of magnitude. We believe that such a behaviour could be physically meaningful, in view of the fact that it is found here for the first time within the wide systematics accumulated so far. It is interesting indeed to remark that predictions of fission-like models like the one of Ref. 9 start to diverge from those given by generalized α-decay models like the one of Ref. 7 for cluster mass $A \sim 30 - 35$.

From the experimental point of view, both the emitted cluster and the mother nucleus have been the heaviest ever studied for this rare kind of radioactivity; moreover, the branching ratio relative to alpha decay is one of the smallest ever measured for hadronic decay modes of nuclei.

2.2. Ne DECAY OF ^{230}U

This experiment [12] aimed at measuring, on the other hand, a quite light cluster, ^{22}Ne, emitted by the lightest isotope of the uranium isotopic series, ^{230}U. In addition to comparison with theoretical predictions, the motivation was here that the large systematics accumulated on uranium isotopes in recent years (A=232,3,4,5,6) [5] pointed out the different behavior of the partial half lives for spontaneous fission and cluster radioactivities with the mass number of the parent, the former being practically constant and the latter rapidly varying as a consequence of shell and other structure effects. In view of the additional feature that both spontaneous fission and cluster radioactivity have, for this particular isotopic series, partial half lives of similar order of magnitude, it was found interesting to extend the systematics both from the light and heavy sides. We will discuss here the measurement of cluster decay of the lightest uranium isotope, ^{230}U, while the one on the heaviest, ^{238}U, is still in progress, as will be discussed later.

The ^{230}U source was obtained, also in this case, by radiochemistry following an intense proton irradiation of metallic thorium targets producing some GBq activity of ^{230}Pa, subsequently β-decaying into ^{230}U. This is a further example of how challenging is to obtain a sufficient number of mother nuclei in the most advanced experiments on exotic decay. The detectors were still of the glass type, this time arranged in 4π geometry. Six events were found in the course of the scanning; subsequent comparison with accelerator calibrations allowed to attribute them to ^{22}Ne clusters. The branching ratio was $(4.8 \pm 2.0) \times 10^{-14}$ and the partial half life was $(3.7 \pm 1.5) \times 10^{19}$ s. Comparison with theoretical predictions shows, this time, a beautiful agreement with predictions of the "α decay"-like model of Ref. 8, and only a fair agreement with the fission-like model of Poenaru [9].

2.3. ^{14}C DECAY OF ^{225}Ac

One of the most important achievements on heavy cluster radioactivity was the discovery of the sensitivity of its partial half life to the microscopic properties of the mother-daughter nuclei. This resulted from many evidences like e.g. the fine structure in the energy spectrum of ^{14}C clusters in the well known ^{223}Ra decay, the hindered decay of odd-A emitters like ^{221}Fr, ^{221}Ra, ^{223}Ra, ^{231}Pa, ^{233}U and others, the anomalously high/low values of two-clusters branching ratios such as ^{24}Ne/^{28}Mg, ^{23}F/^{24}Ne and others [5].

Within such a framework, ^{225}Ac was a special case since while being an odd-Z nucleus its partial decay rate as well as its branching ratio relative to α decay do not seem to exhibit any special hindrance like the one of other odd-A exotic emitters. The situation was summarized in Ref. 13 in the case of hindrance factors for ^{14}C emitters. This quantity, borrowed from α decay, measures the degree according to which an odd-A transition is slower in comparison with an even-even one having the same barrier penetrability. While typical hindrance factors,

$$HF = \frac{\gamma^2(A+1) + \gamma^2(A-1)}{2\gamma^2(A)},$$

where $\gamma^2(A)$ is the reduced width of the cluster emitter of mass number A, are in the range 10-100, the one of the $^{225}\text{Ac} \rightarrow ^{14}\text{C}$ decay measured in 1993 [14] is practically unitary.

The fact that such a case seems surprisingly to behave like an even-even one has been variously justified [14] by using arguments based on the microscopic structure of ^{225}Ac.

In order to have a firmer basis for any theoretical interpretation it was found important to have further experimental data in order, as a first instance, to confirm the 1993 result and possibly to achieve a deeper insight on this rather interesting exotic decay.

In the most recent experiment [15] a new, strong ^{225}Ac source was prepared to be used in two independent set-ups:

1. a high efficiency but low energy resolution experiment to remeasure the integrated decay rate of the $^{225}\text{Ac} \rightarrow ^{14}\text{C} + ^{211}\text{Bi}$ decay;

2. a high resolution experiment to study the energy spectrum of the emitted ^{14}C clusters.

Like in previous experiment, the ^{225}Ac source, $T_{1/2}$ = 10.0 d, was obtained by means of the intense, mass-separated 60 keV ^{225}(Fr+Ra) beam produced at ISOLDE, CERN, where such nuclides are obtained through spallation reactions induced by the 1 GeV proton beam of the PS-Booster on a thick ThC$_2$ target. After extraction, post-acceleration and magnetic separation the ^{225}(Fr+Ra) ions were sent in a collection chamber and implanted onto a vitreous carbon catcher. At this point the ^{225}Ac source was simply obtained by letting it β-decay; again, glass detectors in 2π geometry were used to detect ^{14}C cluster decay.

14 events were found in 86.2 cm^2 glass surface. From the known source intensity and irradiation time, the branching ratio for ^{14}C decay of ^{225}Ac came out to be $B = (4.5 \pm 1.4)10^{-12}$ and the corresponding partial half life $T_{1/2} = (1.9 \pm 0.6)10^{17}$ s, in very good agreement with the 1993 result [14].

One of the goals of the experiment was to find out which level of the ^{211}Bi residual nuclide is preferentially fed by the ^{14}C decay of ^{225}Ac. Two hypothesis have been put forward to explain its unexpectedly high decay rate, in analogy with α decay, in terms of favoured transition either to the ground state or to the first excited one of ^{211}Bi [14]. While theoretical arguments based on the structure of the ^{225}Ac mother nucleus could justify both hypothesis, only an experimental result could solve such ambiguity, thus possibly confirming the use of ^{14}C radioactivity as a spectroscopic tool after the pioneering experiments with ^{223}Ra decay [16].

The ^{225}Ac source was therefore used in a second set-up aimed at measuring the ^{14}C spectrum with an energy resolution sufficient to study its fine structure. The SOLENO spectrometer of IPN-Orsay was used for this purpose. The basic idea is to use the selective features of the spectrometer to reject the high flux of α particles, while keeping the good energy resolution typical of Si detectors.

Unfortunately, the experiment turned out to be difficult due to multiple α pile-up events because of the extremely high intensity of the source which unfortunately fell in the energy region between 28 and 29 MeV where ^{14}C events were expected. It was therefore impossible to unambiguously assign the recorded events to ^{14}C.

However, the positive and consistent result obtained with track detectors confirms the interest of ^{14}C decay of ^{225}Ac from the point of view of nuclear structure. It also suggests that an experimental identification of the favoured transition, a crucial information to test the proposed theoretical interpretations, is a very difficult task indeed.

2.4. EXPERIMENTS IN PROGRESS

We will now discuss two experiments in progress aiming at measuring the cluster decay of ^{238}U by ^{34}Si emission and the one of ^{232}Ac by ^{14}C and ^{15}N emissions. Both experiments have an ultra-decennal life, but are now close to the conclusion.

The former, started in 1995, is being performed by the Dubna-Milano collaboration and was motivated by (i) extending the systematics on uranium isotopes previously discussed in the experiment on ^{230}U towards the heaviest member of the isotopic series, and (ii) finding (or not) a confirmation of the behaviour outlined with ^{34}Si decay of ^{242}Cm, i.e. the prevalence of the "fission-like" mechanism of Poenaru in respect to the "α decay"-like ones for such heavy clusters.

In this experiment 1500 cm^2 of uranium metallic foils kindly obtained on loan from Goodfellow Ltd, Cambridge, UK, were used to irradiate on the 2 sides for 2 years 3000 cm^2 of PET foils acting as track detectors. Detector analysis was very time consuming because of the enormous surface of the detectors and very difficult since a thick source was used, this giving rise to a spread in the track geometrical parameters. Several candidates have been found and are presently being carefully compared with accelerator calibrations.

The experiment on ^{223}Ac was mainly motivated both by structure arguments similar as those discussed in the case of ^{225}Ac and also, and especially, by the fact that it might allow to discover ^{15}N emission, which comes out to be particularly favoured in this case due to shell effects in ^{208}Pb residual nucleus. ^{15}N would be not only another cluster, but, more important, an odd-Z cluster, thus possibly allowing to study even-odd effects in the light cluster itself.

The idea to run this experiment came out in 1993 but only more recently it was possible to find a solution to obtain a strong enough ^{223}Ac source.

The solution was to irradiate by an intense 66 MeV proton beam a thick thorium target thus producing ^{227}Pa ($T_{1/2}$ = 38 min) nuclides by means of the ^{232}Th(p,6n) reaction, doing an on-line chemistry to separate Pa from Th, depositing it on a golden-plated disk and letting it α decay in order to get the wanted ^{223}Ac source. The only laboratory combining a proton beam with the required energy and intensity and on-line radiochemistry was found to be iThemba at Faure, South Africa. The Faure-Dubna-Milano-Moscow collaboration was able to perform three irradiations of glass detectors ensembles in 2005. Detector analysis aiming at measuring both ^{14}C and ^{15}N emissions is almost finished and results will be published quite soon.

3. **CONCLUSIONS**

Table 1 shows all the experimental data available for cluster decay. As one clearly sees, the progress made in this field is impressive since 1984, the starting year of the story.

However it is also clear, as the experiments discussed in previous section have pointed out, that large room is still available for further progress, both from the experimental and (especially) the theoretical side. As a matter of fact, we can mention at least three still open problems, namely the possible existence of other islands of cluster radioactivity in which the residual nuclei are close to other doubly-magic ones like ^{100}Sn or ^{132}Sn, the investigation of even-odd effects and the study of the emission of heavy clusters in view of a better understanding of the theoretical interpretation of this phenomenon.

Table 1

Cluster decay experimental data

Emitter	Cluster	Q(MeV)	Detection System	$B = \lambda_{cl}/\lambda_\alpha$	$\lg_{10} T(s)$
^{114}Ba	^{12}C	18.3-20.5	POLY	$< 10^{-4}$	> 3.63
^{114}Ba	^{12}C	18.3-20.5	BP1	$< 3.4 \cdot 10^{-5}$	> 4.10
^{221}Fr	^{14}C	31.28	BP1	$(8.14 \pm 1.14)10^{-13}$	14.52
^{221}Ra	^{14}C	32.39	BP1	$(1.15 \pm 0.91)10^{-12}$	13.39
^{222}Ra	^{14}C	33.05	POLY	$(3.7 \pm 0.6)10^{-10}$	11.01
^{222}Ra	^{14}C	33.05	SOLENO	$(3.1 \pm 1.0)10^{-10}$	11.09
^{222}Ra	^{14}C	33.05	SOLENO	$(2.3 \pm 0.3)10^{-10}$	11.22
^{223}Ra	^{14}C	31.85	$E \times \Delta E$	$(8.5 \pm 2.5)10^{-10}$	15.06
^{223}Ra	^{14}C	31.85	SOLENO	$(5.5 \pm 2.0)10^{-10}$	15.25
^{223}Ra	^{14}C	31.85	$E \times \Delta E$	$(7.6 \pm 3.0)10^{-10}$	15.11
^{223}Ra	^{14}C	31.85	POLY	$(6.1 \pm 1.0)10^{-10}$	15.20
^{223}Ra	^{14}C	31.85	SPLIT-POLE	$(4.7 \pm 1.3)10^{-10}$	15.32
^{223}Ra	^{14}C	31.85	SOLENO	$(6.4 \pm 0.4)10^{-10}$	15.19
^{223}Ra	^{14}C	31.85	SOLENO	$(7.0 \pm 0.4)10^{-10}$	15.14
^{223}Ra	^{14}C	31.85	SOLENO	$(8.9 \pm 0.4)10^{-10}$	15.04
^{224}Ra	^{14}C	30.54	POLY	$(4.3 \pm 1.2)10^{-11}$	15.86
^{224}Ra	^{14}C	30.54	SOLENO	$(6.5 \pm 1.0)10^{-11}$	15.68
^{225}Ac	^{14}C	30.48	BP1	$(6.0 \pm 1.3)10^{-12}$	17.16
^{225}Ac	^{14}C	30.48	BP1	$(4.5 \pm 1.4)10^{-12}$	17.28
^{226}Ra	^{14}C	28.21	SOLENO	$(3.2 \pm 1.6)10^{-11}$	21.19
^{226}Ra	^{14}C	28.21	POLY	$(2.9 \pm 1.0)10^{-11}$	21.24
^{226}Ra	^{14}C	28.21	POLY	$(2.3 \pm 0.8)10^{-11}$	21.34
^{228}Th	^{20}O	44.72	BP1	$(1.13 \pm 0.22)10^{-13}$	20.72
^{231}Pa	^{23}F	51.84	BP1	$(9.97^{+22.9}_{-8.28}$	26.02
^{230}Th	^{24}Ne	57.78	PET	$(5.6 \pm 1.0)10^{-13}$	24.61
^{232}Th	^{24,26}Ne	55.62,55.97	PET	$< 2.82 \cdot 10^{-12}$	>29.20
^{231}Pa	^{24}Ne	60.42	PET	$6 \cdot 10^{-12}$	23.23

(continues)

Table 1 (continued)

Emitter	Cluster	Q(MeV)	Detection System	$B = \lambda_{cl}/\lambda_\alpha$	$\lg_{10} T(s)$
^{231}Pa	^{24}Ne	60.42	BP1	$(1.34 \pm 0.17)10^{-11}$	22.88
^{230}U	^{22}Ne	61.40	BP1	$(4.8 \pm 2.0)10^{-14}$	19.57
^{232}U	^{24}Ne	62.31	PET	$(2.0 \pm 0.5)10^{-12}$	21.08
^{232}U	^{24}Ne	62.31	PSK50	$(8.68 \pm 0.93)10^{-12}$	20.42
^{232}U	^{24}Ne	62.31	PSK50	$(9.16 \pm 1.10)10^{-12}$	20.40
^{233}U	^{24,25}Ne	60.50,60.75	PET	$(7.5 \pm 2.5)10^{-13}$	24.83
^{233}U	^{24,25}Ne	60.50,60.75	PSK50	$(7.2 \pm 0.9)10^{-13}$	24.84
^{234}U	^{24,26}Ne	58.84,59.47	PSK50	$(9.06 \pm 6.60)10^{-14}$	25.92
^{234}U	^{24,26}Ne	58.84,59.47	PET	$(9.90 \pm 9.90)10^{-14}$	25.88
^{235}U	^{24,25}Ne	57.36,57.83	PET	$(8.06 \pm 4.32)10^{-12}$	27.42
^{236}U	^{24,26}Ne	55.96,56.75	PET	$< 9.2 \cdot 10^{-12}$	>25.90
^{232}U	^{28}Mg	74.32	PSK50	$< 1.18 \cdot 10^{-13}$	>22.26
^{233}U	^{28}Mg	74.24	PSK50	$< 1.30 \cdot 10^{-15}$	>27.59
^{234}U	^{28}Mg	74.13	PET	$(2.3^{+0.8}_{-0.6}$	27.54
^{234}U	^{28}Mg	74.13	PSK50	$(1.38 \pm 0.25)10^{-13}$	25.14
^{235}U	^{28,29}Mg	72.20,72.61	PET	$< 1.8 \cdot 10^{-12}$	>28.09
^{236}U	^{28,30}Mg	71.69,72.51	PET	$2.0 \cdot 10^{-13}$	27.58
^{237}Np	^{30}Mg	75.02	PET	$< 8.0 \pm 10^{-14}$	>26.93
^{237}Np	^{30}Mg	75.02	PSK50	$< 1.8 \cdot 10^{-14}$	>27.57
^{236}Pu	^{28}Mg	79.67	PET	$2.0 \cdot 10^{-14}$	21.67
^{236}Pu	^{28}Mg	79.67	PHOSP. GLASS	$(2.7 \pm 0.7)10^{-14}$	21.52
^{238}Pu	^{28,30}Mg	75.93,77.03	LG750	$(5.62 \pm 3.97)10^{-17}$	25.70
^{238}Pu	^{32}Si	91.21	LG750	$(1.38 \pm 0.50)10^{-16}$	25.27
^{240}Pu	^{34}Si	90.95	PET	$< 6 \cdot 10^{-15}$	>25.52
^{241}Am	^{34}Si	93.84	POLY	$< 2.6 \cdot 10^{-13}$	>22.71
^{241}Am	^{34}Si	93.84	PET	$< 5.4 \cdot 10^{-15}$	>24.41
^{241}Am	^{34}Si	93.84	LG750	$< 7.4 \cdot 10^{-16}$	>25.26
^{242}Cm	^{34}Si	96.53	LG750, GOI-104	10^{-16}	23.15

As far as the first point is concerned, several attempts to measure cluster radioactivity of ^{114}Ba have already been performed [17] leading to the conclusion that a positive result would need a very serious improvement of the experimental conditions which, unfortunately, does not seem to be immediately feasible in view of this extremely out-of-the-line-of-stability nuclide. Moreover, it is hoped that the experiment on ^{223}Ac previously described as well as future renewed attempts to detect the fine structure in the ^{14}C spectrum from ^{225}Ac might allow to clarify even-odd and related nuclear structure effects. Finally, other heavy cluster emissions might be studied, for instance, from ^{241}Am. However, it is clear that the above experimental efforts will be sterile if not supported by an appropriate theoretical interpretation.

We hope that this might happen with the same enthusiasm and effectiveness which characterized the work of Dorin Poenaru in the early years of life of this exciting field of nuclear physics.

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