Abstract

Using on-line mass separation of evaporation residues from heavy-ion induced fusion reactions, the α-decay of very neutron deficient nuclei with $52 \leq Z \leq 55$ -- including the recently identified new isotopes $^{116}$Xe and $^{109}$Te -- and $78 \leq Z \leq 85$ was investigated. The results and the systematics of α-decay energies and reduced widths are discussed.

1. Introduction

Very neutron deficient nuclei in the trans-tin region represent the lightest α-emitters presently known. The occurrence of α-decay in this region is due to an enhancement of the α-decay energies as a consequence of the expected double shell closure at $Z = N = 50$. Experimental Qα-values and reduced α-decay widths allow a first insight into the shape of the mass-energy surface and nuclear structure in the vicinity of the doubly magic nucleus $^{150}$Sn.

Alpha-decay of tellurium isotopes was first observed in 1965 by Macfarlane and Silivola and later by Bogdanov et al. Both groups investigated evaporation residues from $^{189}$Ru($^{10}$O,xn) reactions using a helium-jet transport system. A variety of very neutron deficient trans-tin isotopes was found in fusion reactions of the type $^{58}$Ni($^{4}$Ni,xyn), making use of high-intensity beams of heavy ions which became available at the UNILAC in Darmstadt about five years ago. The wide distribution of residual nuclei and their complex decay behaviour ($\beta^+/\beta^-$-decay, ground-state α-decay, $\beta$-delayed proton and α-emission) makes selective detection technique indispensable. On-line mass separation combined with α-spectroscopy and time-correlation techniques proved to be a powerful method, leading to the identification of fourteen isotopes which form a new island of α-emission.

The region around $Z = 82$ has attracted much interest since the observation of the odd-even staggering of nuclear charge radii in the light mercury isotopes $^{198}$Hg. The pronounced odd-even effect exhibited by both α-decay energy and reduced width of these isotopes is probably closely connected with the systematic change of ground-state shapes between the odd and the even isotopes. Our experiments on the α-decay of very neutron deficient nuclei with $78 \leq Z \leq 85$, especially the studies of lead and mercury decays, should help to gain more information on nuclear structure in this region.

2. Experimental techniques

The experiments were performed at the GSI on-line mass separator connected to the heavy-ion accelerator UNILAC. Very neutron deficient trans-tin isotopes were produced as evaporation residues from reactions of 5 MeV/u $^{58}$Ni ions on $^{58}$Ni targets with a thickness of 4 mg/cm$^2$. For the measurements in the lead region, enriched targets of $^{132,134}$Xe and $^{107}$Ag were bombarded with 4.6-6.3 MeV/u beams of $^{58}$Ni and $^{58}$Kr. The recoiling reaction products were stopped in a catcher (kept at a temperature of about 2400 °C), reionized, mass separated, and investigated by α-spectroscopy. Using a modified version of the FEBIAD ion source with a reduced volume of 2 cm$^3$, an intensity of, for example, 120 atoms/s was obtained for the $^{110}$I beam. The thermal ion source used for the $^{110}$Cs measurements yielded an intensity of 700 atoms/s for this isotope.

Two arrays of surface-barrier detector telescopes served for measuring proton- and α-decays (for details see Ref. 5): Half-life and parent-daughter correlation measurements were carried out with two telescopes face to face at one of the counting positions of a 'windmill' system. For "in-beam" measurements the mass-separated ion beam was implanted into a thin carbon foil (12 μg/cm$^2$) behind which a telescope was placed. In order to allow efficient time correlation measurements for short half-lives, a 50 μm thick annular detector was put in close geometry in front of the collector foil, the ion beam passing through the central hole of the detector.

The linear signals from ΔE and E detectors were stored event by event on magnetic tape by a PDP 11/45 computer. Discrimination between α-particles and $\beta$-delayed protons which occur in the same energy region was obtained by analysing ΔE signals in anticoincidence or coincidence with E signals. With each event the real time given by a 100 kHz driven scaler was registered.

3. Experimental results and discussion

3.1 Trans-tin region

The results from α-decay measurements are compiled in Table 1. They include the recently identified new isotopes $^{116}$Xe and $^{106}$Te as well as improved decay data (compared to those of Ref. 5) for $^{198,199}$Te, $^{198,199,200}$I, $^{111,112}$Xe, and $^{117}$Cs. The new results will be discussed in the following.
Table 1

Summary of α-decay measurements in the trans-stin region

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>T_{1/2} a) (s)</th>
<th>E_α (keV)</th>
<th>P_{g.st.α} b)</th>
<th>W_α</th>
</tr>
</thead>
<tbody>
<tr>
<td>110mTe</td>
<td>(60 ± 10) × 10^{-6}</td>
<td>4160 ± 30</td>
<td>0.7 ± 0.3 d)</td>
<td>3.7 ± 1.6</td>
</tr>
<tr>
<td>118mTe</td>
<td>(3.6 ± 1.8) × 10^{-3}</td>
<td>3833 ± 15 d)</td>
<td>0.68 ± 0.12 d)</td>
<td>1.7 ± 0.7</td>
</tr>
<tr>
<td>118Te</td>
<td>2.1 ± 0.1</td>
<td>3320 ± 20 e)</td>
<td>[3.9 ± 1.3 × 10^{-2}]</td>
<td>2.8 ± 1.0</td>
</tr>
<tr>
<td>116mTe</td>
<td>4.1 ± 0.2</td>
<td>3080 ± 15 d)</td>
<td>[7.6 × 10^{-4}]</td>
<td>3.8 ± 1.8</td>
</tr>
<tr>
<td></td>
<td>18.6 ± 0.8</td>
<td>2624 ± 15 e)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>110I</td>
<td>0.65 ± 0.02</td>
<td>3444 ± 10 e)</td>
<td>0.17 ± 0.04</td>
<td>1.5 ± 0.5</td>
</tr>
<tr>
<td>111I</td>
<td>2.5 ± 0.2</td>
<td>3152 ± 10 d)</td>
<td>[4.6 × 10^{-3}]</td>
<td></td>
</tr>
<tr>
<td>112I</td>
<td>3.42 ± 0.11</td>
<td>2880 ± 30 e)</td>
<td>[5.0 × 10^{-5}]</td>
<td></td>
</tr>
<tr>
<td>113I</td>
<td>5.9 ± 0.5</td>
<td>2610 ± 40 e)</td>
<td>[5.3 × 10^{-7}]</td>
<td></td>
</tr>
<tr>
<td>116Xe</td>
<td>0.2 f)</td>
<td>3737 ± 30</td>
<td></td>
<td></td>
</tr>
<tr>
<td>112Xe</td>
<td>0.74 ± 0.20</td>
<td>3480 ± 30 e)</td>
<td>[0.5] f)</td>
<td></td>
</tr>
<tr>
<td>112Xe</td>
<td>0.89 ± 0.20</td>
<td>3580 ± 30 e)</td>
<td>[0.048] f)</td>
<td></td>
</tr>
<tr>
<td>113Xe</td>
<td>2.7 ± 0.8</td>
<td>3210 ± 20 e)</td>
<td>[0.26] f)</td>
<td></td>
</tr>
<tr>
<td>114Xe</td>
<td>2.8 ± 0.2</td>
<td>2985 ± 15 d)</td>
<td>[2.9 × 10^{-3}]</td>
<td></td>
</tr>
<tr>
<td>114Cs</td>
<td>0.57 ± 0.02 g)</td>
<td>3239 ± 30 e)</td>
<td>[5.9 × 10^{-5}]</td>
<td></td>
</tr>
</tbody>
</table>

a) From Ref. 5 except for the nuclides 116mTe, 115I, 114Xe, 111Xe and 114Cs.
b) Values in square brackets are from alpha-particle transmission calculations according to Rasmussen (Ref. 14), assuming W_{g.s.} = 1.
c) As the determination of the branching ratio via parent-daughter relationships is not accurate, the predominance of alpha-decay P_{g.st.α} ≈ 1 was deduced from a comparison between the measured half-life and the predicted value of 1 s from the gross theory of beta decay (Refs. 15, 16).
d) From Ref. 5.
e) Remeasurements with generally improved precision compared to those of Ref. 5.
f) Assuming equal decay probabilities for alpha and beta decay, the half-life predicted for the latter being 0.4 s from the gross theory of beta decay (Refs. 15, 16).
g) From Ref. 17.

At mass 110 the improved counting statistics allowed a further investigation of the two α-lines which appear in the α-spectrum shown in Fig. 1 above the 115I α-line, and which had already been preliminarily assigned to the decays of 116Xe and 118Te. The latter assignment could be confirmed by a time-correlation analysis. Using the in-beam detector system with the additional annular detector as described in Section 2, the time differences between the events in the 3737 keV line and the first following events in the 4160 keV line were analysed. The resulting distribution of time differences is shown in Fig. 2. It includes "cross correlations" between the two opposite detectors as well as "self correlation", that means detection of correlated events in one and the same detector. The occurrence of truly correlated events, showing up at time distances which are 10⁶ times shorter than the mean time distance of the random distribution, clearly proves the parent-daughter relationship of the two lines. From the correlation peak the half-life of 116mTe was determined to be 60 ± 10 μs.

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Fig. 1 Alpha spectrum obtained at mass 110 from the α detector in anticoincidence with the Ε detector of the in-beam telescope. The counting time amounted to 25 h with an average 59Ni beam intensity of 1.6 × 10¹¹ s⁻¹. The positions of possible fine structure in the decay of 115I are indicated by arrows.
The $\alpha$-energies for $^{110}Te$ and $^{111}I$ are in agreement with earlier measurements\textsuperscript{5} but could be determined with higher precision. Even though the $^{111}I$ $\alpha$-line contains about 10$^4$ counts, there is no clear evidence for fine structure in this decay. The branching ratio $F_{\alpha, 106} = 0.17 \pm 0.04$ for ground-state $\alpha$-decay of $^{110}I$ was derived from a new measurement with the in-beam detector at mass 114, where $^{110}I$ is observed as $\alpha$-decay daughter of $^{114}Cs$ (Fig. 3). The measured intensity of $^{111}I$ $\alpha$-decays was corrected by 25% for the loss of activity due to the recoil range in the collector foil exceeding the implantation depth. The non-observation of the $^{114}Xe$ $\alpha$-decay, which is expected on systematical grounds to occur at an energy of about 2.8 MeV (see Fig. 3), results in an upper limit of about 10$^{-6}$ for the $\alpha$-branching ratio.

At mass 114 a 9.7 h measurement was performed with the in-beam telescope, resulting in more precise values for the $\alpha$-energies and half-lives for the $^{114}Xe$ $\alpha$-lines at 3480 keV and 3580 keV. The low-energy member of the doublet is now free from the $^{111}I$ contamination which had obscured this line in earlier experiments\textsuperscript{9}. Both $\alpha$-lines had been shown to be time-correlated with the decay of the 3.6 ms $^{107}Te$ daughter\textsuperscript{7}). Their half-lives were measured by periodically interrupting the mass-separated ion-beam using an electrostatic deflector device. Since the values of 0.74 $\pm$ 0.20 s for the 3480 keV $\alpha$-line and 0.89 $\pm$ 0.20 s for the 3580 keV $\alpha$-line are not significantly different, it still remains unclear whether the two transitions arise from isomerism in $^{114}Xe$ or the feeding of two final states in $^{107}Te$.

A discussion of the experimental results presented in Table 1 within the systematics of $Q_\alpha$-values and reduced decay widths, is of interest for testing mass-formulae predictions in a region far off from stability and for predicting the properties of neighbouring, yet unknown, isotopes. However, one has to keep in mind some restrictions. There is no experimental evidence so far on whether the observed $\alpha$-transitions connect two ground states or whether isomeric and excited states are involved. Furthermore, the systematic behaviour of mass differences ($Q_\alpha$-values) permits only an indirect test of mass formulae. On the other hand, one can obtain masses for far-unstable nuclei (e.g. $^{114}Cs$, $^{111}I$, $^{105}Sb$) by linking $Q_\alpha$-values to known mass excesses\textsuperscript{19,20}.

A comparison of the measured $Q_\alpha$-values with the predictions from mass formulae\textsuperscript{21,22} gives best overall agreement, with a standard deviation of about 0.2 MeV, with the droplet-model of Myers and of Hilf et al. (see Fig. 4). This agreement has an interesting aspect. A decomposition of the theoretical $Q_\alpha$-values into a droplet and a shell-correction part shows that for the $\alpha$ isotopes, for example, about 90% of the $Q_\alpha$-value arises from the shell-correction term. Therefore the agreement with the experimental data within 0.2 MeV is a strong indication for the expected double-shell closure at $Z = N = 50$.

Macfarlane and Siivola\textsuperscript{13} pointed out that a kind of "superallowed" $\alpha$-decay, resulting in a large reduced $\alpha$-width, might be observed for very neutron deficient Te isotopes which have their "valence" neutrons and protons in the same single-particle level. In order to calculate reduced $\alpha$-widths, one has to know the energy, the half-life and the branching ratio of the decay. The reduced widths $W_{\alpha}$ given in Table 1 were calculated according to the formula $W_{\alpha} = \lambda_{\alpha} P$, where $\lambda_{\alpha}$ is the partial transition probability for $\alpha$-decay and $P$ the barrier penetrability for $\alpha$-particles\textsuperscript{13,15}. Comparing the $W_{\alpha}$ values of the even-even emitters $^{104}Te$ and $^{106}Te$ with the body of existing data for heavier nuclei (Fig. 5)\textsuperscript{5,23,27}, it can be seen that they follow the general trend of the systematics, i.e. they increase with decreasing neutron number between two shell closures. There seems to be no evidence for an additional enhancement of $W_{\alpha}$ for the tellurium isotopes.
3.2 Lead region

In the lead region $\alpha$-energy spectra obtained from mass-separated samples are generally more complex than those in the trans-tin region. The assignments of $\alpha$-energies to specific nuclei are based on the mass number, determined unambiguously by the magnetic separation, and on $\alpha$-decay systematics. The $\alpha$-spectrum measured at mass number 183 (Fig. 6) may serve as an example. At lower energies, one observes the known $\alpha$-lines of $^{196}$Au, and of $^{195}$Hg and its $\alpha$-decay daughter $^{192}$Pt. The new lines occurring at higher energies are then assigned to higher-Z isotopes, where the highest possible $Z$-value is given by the projectile-target combination -- this case

82. Supported by the $\alpha$-energy systematics for thallium and lead, we assigned the $\alpha$-lines at 6343, 6378 and 6449 keV to $^{183}$Tl (confirming earlier unpublished data\cite{19}) and those at 6715 and 6798 keV to $^{183}$Pb. These assignments have been further confirmed by producing $^{183}$Tl instead of $^{185}$Pb as compound nucleus. The lines at 6715 keV and 6798 keV disappeared while the other lines were still observed. In a similar way $\alpha$-lines of 6632 keV and 6820 keV from reactions of $^{196}$Tl on $^{193}$Nd and of $^{195}$Kr on $^{197}$Ag were assigned to $^{183}$Pb and $^{183}$Tl, respectively. The decay properties of the new isotopes are summarized in Table 2.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$E_\alpha$ (keV)</th>
<th>I $^a$ (I)</th>
<th>$T_{1/2}$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{188}$Bi</td>
<td>6820 ± 20</td>
<td>85 ± 9</td>
<td></td>
</tr>
<tr>
<td></td>
<td>7005 ± 25</td>
<td>15 ± 9</td>
<td></td>
</tr>
<tr>
<td>$^{183}$Pb</td>
<td>6715 ± 20</td>
<td>92 ± 4</td>
<td>0.6 ± 0.3</td>
</tr>
<tr>
<td></td>
<td>6798 ± 25</td>
<td>8 ± 4</td>
<td></td>
</tr>
<tr>
<td>$^{184}$Pb</td>
<td>6632 ± 10</td>
<td></td>
<td>0.55 ± 0.06</td>
</tr>
<tr>
<td>$^{183}$Tl</td>
<td>6343 ± 10</td>
<td>83 ± 4</td>
<td>0.053 ± 0.004</td>
</tr>
<tr>
<td></td>
<td>6378 ± 15</td>
<td>16 ± 2</td>
<td>0.052 ± 0.006</td>
</tr>
<tr>
<td></td>
<td>6449 ± 15</td>
<td>1.0 ± 0.3</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Relative intensities
The surprisingly short half-life observed for $^{183}$Tl might be explained if we assign it to an isomeric $\frac{3}{2}^-$ state which is de-excited predominantly by an E3-transition to a $\frac{5}{2}^-$ state. On the basis of systematics of $\frac{3}{2}^-$ and $\frac{5}{2}^-$ levels in $^{183-187}$Tl [26], the expected lifetime of the $\frac{3}{2}^-$ isomer would be of the order of the measured one.

For $^{185-186}$Pb and $^{184,185,187}$Tl, the existing data [25] were confirmed, except for $^{185}$Pb, where the measured half-life of 4.7 $\pm$ 1.1 ms is considerably shorter than the value of 7.9 $\pm$ 1.6 s given by Gauvin et al. [29]. A shorter half-life in turn increases the reduced $\alpha$-width for this isotope [30].

For $^{182-186}$Hg and $^{178,179}$Pt, $\alpha$-branching ratios were determined from comparison of the intensities of parent and daughter activities in the same spectrum (see Fig. 6). Our results for $^{178,179}$Pt and $^{185}$Hg agree with the literature values [31] whereas the values for $^{183,184}$Hg deviate substantially from those given in Ref. 35. The literature values were deduced from a comparison of the intensities of K X-rays and $\alpha$'s and include an estimated correction for the contribution of K X-rays due to internal conversion. The uncertainty in the correction term in cases where the decays are not sufficiently known probably explains the observed discrepancy.

The reduced $\alpha$-decay widths derived from $\alpha$-energies and half-lives from Refs. 9 and 35, and the new $\alpha$-branching ratios for $^{182,183,184}$Hg are $0.8 \pm 0.1$, $1.0 \pm 0.1$ and $0.6 \pm 0.2$, respectively [11]. The behaviour of reduced widths of $\alpha$ isomers was earlier discussed by Horwitz et al. [39] and Hagberg et al. [40]. The revised width for $^{182}$Hg means that for the even isotopes a regular behaviour in $W_\alpha$ is established when going from $^{182}$Hg to $^{184}$Hg (see Fig. 5). The effect of nuclear deformation on the barrier transmission in $\alpha$-decay was discussed by Rasmussen and Segall [59]. Since this effect was not included in our calculation of the penetrability, it appears likely that the observation of larger widths for $^{184,185}$Hg as compared to their even neighbors is related to the ground-state deformation of these nuclei.

### 3.3 Search for proton radioactivity

With $^{113}$Cs and $^{109}$I one has reached the last proton stable isotopes of these elements according to the predictions of most current mass formulae [82]. The high yields for such extremely neutron deficient isotopes obtained from heavy-ion reactions (see Section 2) offer good conditions for search experiments aimed at finding proton decay from the ground state. In this region of medium-mass nuclei a proton-decay energy of a few hundred keV, which could be gained by just crossing the drip line, would be sufficient to give a detectable decay branch. However, the main experimental problem in the direct detection of proton radioactivity lies in the small "observation" window [83] defined by the lowest detectable branch on one side and by the shortest detectable half-life on the other side. Owing to the lower and narrower Coulomb barrier for protons, this window is much smaller than for $\alpha$-decay. D'Anzio et al. [61] performed a search for $^{113}$Cs at the ISOLDE mass separator and estimated the observation window to $540 < E_\beta < 650$ keV, corresponding to the conditions $P_\beta > 10^4$ and $T_\beta > 5$ ms. [60]

All attempts to detect proton radioactivity from $^{113}$Cs and $^{109}$I remained unsuccessful so far. However, the non-observation of any activity of $^{113}$Cs points to a considerable half-life decrease due to proton-decay [85-87]. In the case of $^{109}$I the short (80 ms) partial half-life for ground state $\alpha$-decay, estimated from systematics [10] could just as well explain the non-observation. For antimony one might expect an additional effect due to the $Z = 50$ shell closure which could move the drip line somewhat towards higher mass numbers. The result of an alpha-proton correlation study for the decay of $^{113}$I+$^{109}$Sb was negative, however.

For heavy elements such as bismuth, proton-decay energies of approximately 1 MeV are required to give an observable decay branch. Thus, although the new isotope $^{188}$Bi lies beyond the predicted proton drip line, its possible proton-decay branch is presumably far too small to be detected. The main difficulty in the production of very neutron deficient isotopes in this region is found in the increased competition of fission in the de-excitation of the compound nucleus. One can hope, however, that further developments of heavy-ion-based on-line systems such as mass separators and velocity filters will offer good chances for the possible study of ground-state proton emission. The recent work by Hofmann et al. [88] indeed gives support to this optimism.

**References**

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