DECAY PROPERTIES OF NEUTRON-RICH NUCLEI IN THE MASS REGION A=100

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Abstract

Neutron-rich nuclei in the mass region A=100 have been investigated by measurement of γ singles and γ-γ coincidence spectra. The activities have been produced by thermal-neutron induced fission of 235U, 238U and 244Cf using automated chemical procedures for the separation of Y, Zr, Nb, Mo, Tc or Ru from the fission product mixtures. The systematic trends in the low-lying states of even-even nuclei in this region have been obtained and are compared with theoretical interpretations.

As has been pointed out by Arseniev et al.1) as well as in other theoretical studies2-4), neutron-rich nuclei in the mass region A=100 may constitute a new region of deformation. But, in contrast to the nuclei in the rare-earth or actinide region it has been shown that these nuclei are softer on both beta and gamma deformations. This has been demonstrated in form of the potential energy surfaces where in some cases two minima, representing two different shapes of the nucleus, occur. However, both minima are often only slightly different in energy and the higher minimum is not a true one, but one can reach the lower minimum without any significant energy barrier. According to the calculations of Gneuss and Greiner5), the softness in the gamma direction, leading to a lowering of the gamma bands, is always accompanied by a more or less destruction of the rotational structure. Apparently the rotation-vibration interactions in very soft gamma dependent potentials are effective to such an extent that pure rotational bands cannot occur. In other cases, where the potential energy surface shows two minima at different deformations, i.e., softness in the beta direction, low-lying excited 0+ states can be observed. While

Table 1

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Strongest γ-rays [keV]</th>
<th>Nuclide</th>
<th>Half-life</th>
<th>Strongest γ-rays [keV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Y-96</td>
<td>9.60±0.3 sec</td>
<td>617.7, 914.6, 1106.6, 1750.3;</td>
<td>Tc-103</td>
<td>54.2±0.8 sec</td>
<td>136.1, 210.2, 346.3, 562.8;</td>
</tr>
<tr>
<td>Y-97</td>
<td>1.50±0.3 sec</td>
<td>161.4, 1102.9;</td>
<td>Tc-104</td>
<td>18.5±0.5 min</td>
<td>357.8, 530.6, 535.2;</td>
</tr>
<tr>
<td></td>
<td>3.60±0.3 sec</td>
<td>1290.8, 1399.4, 3287.5, 3401.2;</td>
<td>Tc-105</td>
<td>7.6±0.1 min</td>
<td>188.0, 143.2, 159.3, 321.5;</td>
</tr>
<tr>
<td>Zr-99</td>
<td>2.0±0.2 sec</td>
<td>387.6, 469.2, 546.2, 594.1;</td>
<td>Tc-106</td>
<td>36.0±1.0 sec</td>
<td>270.3, 522.4, 720.8, 792.7;</td>
</tr>
<tr>
<td>Zr-100</td>
<td>7.1±0.4 sec</td>
<td>401.0, 504.6;</td>
<td>Tc-107</td>
<td>21.0±1.0 sec</td>
<td>102.7, 160.3, 177.2, 458.9;</td>
</tr>
<tr>
<td>Zr-101 a)</td>
<td>2.0±0.3 sec</td>
<td>-</td>
<td>Tc-108</td>
<td>5.0±0.2 sec</td>
<td>242.4, 465.8, 708.1, 732.4;</td>
</tr>
<tr>
<td>Zr-102 a)</td>
<td>2.9±0.2 sec</td>
<td>-</td>
<td>Tc-109 a)</td>
<td>1.4±0.4 sec</td>
<td>-</td>
</tr>
<tr>
<td>Nb-100</td>
<td>1.5±0.2 sec</td>
<td>159.3, 528.1, 535.4, 1022.1;</td>
<td>Tc-110</td>
<td>1.0±0.2 sec</td>
<td>240.8;</td>
</tr>
<tr>
<td>Nb-101</td>
<td>7.1±0.3 sec</td>
<td>535.4, 600.2, 666.2, 1280.4;</td>
<td>Ru-107</td>
<td>3.8±0.1 min</td>
<td>194.1, 374.5, 462.7, 847.8;</td>
</tr>
<tr>
<td>Nb-102</td>
<td>1.3±0.2 sec</td>
<td>296.0, 400.6, 551.6, 847.6;</td>
<td>Ru-108</td>
<td>6.6±0.1 min</td>
<td>165.1;</td>
</tr>
<tr>
<td>Nb-103</td>
<td>1.5±0.2 sec</td>
<td>296.0, 447.0, 1235.4, 1632.7;</td>
<td>Ru-109</td>
<td>34.5±1.0 sec</td>
<td>206.2, 225.9, 358.7, 1928.9;</td>
</tr>
<tr>
<td>Nb-104</td>
<td>0.8±0.2 sec</td>
<td>192.2, 368.5, 477.4, 812.5;</td>
<td>Ru-110</td>
<td>12.6±0.5 sec</td>
<td>95.8, 112.1;</td>
</tr>
<tr>
<td>Nb-106</td>
<td>-1 sec</td>
<td>171.7;</td>
<td>Ru-111 a)</td>
<td>3 ±1 sec</td>
<td>-</td>
</tr>
<tr>
<td>Mo-103</td>
<td>68.0±1.0 sec</td>
<td>150.2, 424.0;</td>
<td>Ru-112 a)</td>
<td>3.6±0.5 sec</td>
<td>-</td>
</tr>
<tr>
<td>Mo-104</td>
<td>60.0±2.0 sec</td>
<td>66.6, 91.0, 375.8, 420.8;</td>
<td>Ru-113</td>
<td>3.0±0.7 sec</td>
<td>303.6;</td>
</tr>
<tr>
<td>Mo-105</td>
<td>36.0±2.0 sec</td>
<td>64.2, 85.6, 249.5;</td>
<td>Rh-108</td>
<td>17 ±1 sec</td>
<td>434.1, 497.1, 618.9;</td>
</tr>
<tr>
<td>Mo-106</td>
<td>8.2±1.0 sec</td>
<td>53.9, 465.7, 595.3, 618.6;</td>
<td>Rh-109</td>
<td>79.8±1.0 sec</td>
<td>178.0, 291.4, 326.7, 426.1;</td>
</tr>
<tr>
<td>Mo-107</td>
<td>3.5±0.5 sec</td>
<td>64.4, 400.1, 483.3;</td>
<td>Rh-110</td>
<td>3.3±0.3 sec</td>
<td>373.7, 439.7;</td>
</tr>
<tr>
<td></td>
<td></td>
<td>-</td>
<td>Rh-111</td>
<td>11 ±1 sec</td>
<td>275.3;</td>
</tr>
<tr>
<td></td>
<td></td>
<td>-</td>
<td>Rh-112</td>
<td>&lt;1.5 sec</td>
<td>348.6;</td>
</tr>
</tbody>
</table>

a) Mass assignment via genetic relationships with known daughter nuclides

- 483 -
the ground state is represented by the minimum at the spherical point, the second minimum is responsible for the excited $0^+$ state. However, there is no evidence for a rotational band built upon this state. The reason for this is the rather high zero point energy of the spherical ground state. The excited states exceed therefore the barrier between both minima and their wave functions are not longer concentrated in one pot only; they are of mixed character.

From the experimental point of view, studies in the $A=100$ region are hampered by the facts that most of the nuclei of interest are far away from the stability line and have rather short half-lives, and that fission of heavy elements is the only way to produce them.

Our approach to obtain more insight into the properties of these nuclei consists in a combination of rapid chemical separation techniques with high-resolution $\gamma$-ray spectroscopy. We have worked out separation methods for $Y$, Zr, Nb, Mo, Tc and Ru and applied these techniques to the identification of short-lived isotopes of these elements. The nuclides investigated together with their main decay properties are summarized in Table I.

The nuclei have been produced by thermal-neutron induced fission of $^{239}$Pu, $^{235}$Pu and $^{240}$Cf through irradiations performed in a rabbit system of the Mainz Triga reactor delivering in the pulse mode $3 \times 10^{13}$ n/cm$^2$ during 60 msec. The nuclides were isolated from the fission product mixture by rapid chemical separation methods as outlined elsewhere. Conventional $\gamma$-singles and $\gamma-\gamma$ coincidence techniques were applied to reveal the decay schemes.

As an example, some results for neutron-rich technetium and niobium isotopes will be presented. In addition to the neutron-rich technetium iso-

![Image](https://via.placeholder.com/150)

**Fig. 1.** (a) Section of the $\gamma$-ray spectrum (inset) and decay curve of the doublet at 240-242 keV in technetium samples. (b) Growth-and-decay curve of the 206 keV peak of $^{109}$Ru in technetium samples.

![Image](https://via.placeholder.com/150)

**Fig. 2.** Spectrum of $\gamma$-rays of short-lived niobium isotopes from thermal-neutron induced fission of $^{239}$Pu.
fission of $^{252}\text{Cf}$, the $2^+\rightarrow 0^+$ transition in $^{110}\text{Ru}$ has an energy of 260.8 keV. In the γ-ray spectra of the technetium fraction a rapidly decaying peak at this energy has been found located close to the strongest peak of 5.0-sec $^{188}\text{Tc}$ at 242.4 keV as it is shown in the inset of Fig. 1 (a). The decay curve of this doublet shows two components with 5.0±0.5 and 1.0±0.2 sec half-lives. The former half-life belongs to $^{188}\text{Tc}$ whereas the 1.0-sec component is assigned to the decay of $^{110}\text{Tc}$.

Since no γ-rays could be attributed unambiguously to the decay of $^{188}\text{Tc}$, the half-life was determined from the growth-and-decay curves of its known daughter product, 34.5-sec $^{109}\text{Ru}$. Fig. 1 (b) shows the growth-and-decay curve of the strongest γ line at 206 keV. From this curve a half-life of 1.4±0.4 sec for the parent $^{109}\text{Tc}$ is deduced.

For the separation of niobium from the fission product mixture a method has been developed which allows the start of counting 2.2 sec after the end of irradiation5). A γ-ray spectrum of short-lived niobium isotopes covering the energy region up to 2 MeV is shown in Fig. 2. Contributions of longer-lived isotopes have been subtracted. Most of the γ-rays can be attributed to niobium nuclides in the mass region A=99-104. Apart from the well-known peaks of 15-sec $^{99}\text{Nb}$ at 97.4 and 137.5 keV, one can observe γ-rays of two isomers of $^{100}\text{Nb}$ ($t_{1/2}=1.5$ and 3.1 sec) at 159, 528, 538, 600, 1280 keV, the strongest peak of the 7.1-sec $^{101}\text{Nb}$ at 276 keV, the peaks of two isomers of $^{102}\text{Nb}$ ($t_{1/2}=1.3$ and 4.3 sec) at 296, 401, 447, 551, 847, 1633 keV, the 1.5-sec $^{103}\text{Nb}$ with a peak at 103 keV, and γ-rays at 192 and 368 keV belonging to the decay of two isomers of $^{104}\text{Nb}$ ($t_{1/2}=0.8$ and 4.8 sec).

As an example for the data obtained Fig. 3 shows decay curves for $^{100}\text{Nb}$. The 535 keV peak of $^{100}\text{Nb}$, representing the $2^+\rightarrow 0^+$ transition in $^{100}\text{Mo}$, has been observed in both niobium as well as zirconium fractions separated from the fission product mixture. From the decay curve obtained after the separation of niobium the two isomers of $^{100}\text{Nb}$ with 1.5 and 3.1 sec half-lives can be unfolded (Fig. 3a). In the zirconium fraction, however, the growth-and-decay curve (Fig. 3c) of the 535 keV peak showed, after passing the maximum, a decay with 7.1 sec half-life. Hence, the 7.1-sec activity is assigned to $^{100}\text{Zr}$ which decays into a shorter-lived niobium daughter. From the growth of the 535 keV γ line in the zirconium fraction, a half-life of 1.4 sec for the niobium daughter nucleus is deduced, in agreement with the shorter-lived component appearing in Fig. 3a. As the J$^m$ = 0$^+$ ground state of the even nuclei $^{100}\text{Zr}$ should decay preferably to low-spin states in $^{100}\text{Nb}$, the 1.5-sec half-life is assigned to a low-spin isomer of $^{100}\text{Nb}$.

As a result of our investigations the systematic behaviour of the low-lying excited states of neutron-rich even-mass ruthenium and palladium isotopes will be presented.

For the even molybdenum nuclei in the mass region 94-106 (Fig. 4) a steep decrease in the energy of the first excited state was observed with a relatively big jump between $^{100}\text{Mo}$ and $^{102}\text{Mo}$ and a somewhat smaller between $^{102}\text{Mo}$ and $^{104}\text{Mo}$. Simultaneously the 4$^+$ levels shift downwards. The E4/E2 ratio increases without reaching, however, the value of 5.3 expected for a good rotator. A very strange behaviour shows the first excited 6$^+$ state, which is even lower in energy than the first 2$^+$ state in $^{99}\text{Mo}$.

In the calculations of Gneuss and Greiner it has been shown that the spectrum of low-lying states in $^{99}\text{Mo}$ might be explained with a potential energy surface having two minima separated only by a relatively flat barrier. One minimum, corresponding to the ground state, occurs for the spherical shape, the other one, responsible for the low-lying excited

$E_{\gamma_{1+}}$ = 1.81 2.09 1.92 2.12 2.51 2.92 3.04
$E_{\gamma_{2+}}$ = 5973.5 6002.0 6072.4 5956.1 6156.4 6242.3 6433.2 6563.1
$E_{\gamma_{3+}}$ = 6477.2 6753.7 6736.0 6823.1 6991.7 7065.0 7250.0 7275.4
$E_{\gamma_{4+}}$ = 7324.5 7862.7 7923.8 7976.0 7951.7 7920.4 7900.3 7871.2
$E_{\gamma_{5+}}$ = 7945.2 7900.2 7871.2 7842.3 7813.4 7784.5 7755.6 7726.7
$E_{\gamma_{6+}}$ = 7707.1 7679.3 7653.5 7623.7 7594.0 7564.3 7534.6 7504.9
$E_{\gamma_{7+}}$ = 7474.9 7442.2 7410.9 7379.7 7348.5 7317.3 7285.9 7254.4
$E_{\gamma_{8+}}$ = 7224.2 7193.9 7163.5 7133.9 7104.3 7074.7 7045.2 7015.7

Fig. 3. Decay and growth-and-decay curves of the 535 keV (a,c) and 600 keV peak of $^{100}\text{Nb}$ in niobium (a,b) and zirconium samples (c).

Fig. 4. Low-lying levels of even molybdenum nuclei. The data for $^{100}\text{Mo}$ are from this work, for $^{99}\text{Mo}$ and $^{98}\text{Mo}$ from Ref. 8, and for $^{97}\text{Mo}$ from Ref. 9.)
0+ state, is situated in the γ-plane at a deformation of ε=0.45. Sheline et al.,10 have suggested that an inversion of the two minima occurs in heavier molybdenum isotopes: the deformed shape, now, represents the ground state, and the spherical minimum the excited 0+ state. This assumption of shape isomorism seems to be in contradiction with the experimental results of Bohn et al.,11, who have found that the 0+ states in 106Mo and 108Mo decay by fast E2 transitions to the first 2+ states with B(E2) values of 14 single particle units. They have proposed the excited 0+ states in 106,108Mo as band heads for a γ-vibrational band.

On the other hand, as it is known from the rare-earth region, γ-vibrational bands are excited in two-neutron transfer reactions only with a few percent of the ground state strength, while Casten et al.,12 have found in the 109Mo(t,p)110Mo reaction a very high cross section of 52% of the ground state strength for the excitation of the 0+ state in 110Mo at 697 keV. Obviously, the structure of the excited 0+ states in neutron-rich Mo isotopes is more complex than assumed, and more experimental and theoretical investigations are necessary to clear up, for instance, a possible contribution of pairing vibrations to these states.

According to Fig. 5, the energies of the first excited 2+ states in even ruthenium nuclei decrease with increasing neutron number asymptotically towards an energy of about 240 keV for very neutron-rich isotopes. Furthermore, the ratio E2/Eγ of the energies of the first 4+ and 2+ levels increase towards a constant value of about 2.75, far below the value of 3.2 expected for a good rotator. The energies of the second 2+ state also decrease with increasing mass number, as well as that of possible 3+ states which might be interpreted as the first members of γ- or quasi-γ-bands.

A comparison of the 3+ level energies of 106,108,110Ru with the predictions of the asymmetric rotator model of Davydov and Filippov13 leads to good agreements with the observed energies. Asymmetric deformations with a flat minimum at γ = 300° for the neutron-rich Ru isotopes have also been obtained in the calculations of Faessler et al.14).

![Table](image)

Fig. 6. Low-lying levels of even palladium nuclei. The data for 108,110Pd are from this work, for 106,108Pd from Ref.9, and for 112Pd from Ref.7,12.)

Of particular interest is again the behaviour of the excited 0+ states. While in the lighter Ru isotopes the first excited 0+ state can be identified with the third member of the two-phonon triplet, it crosses the other two members in 108Ru and shifts more away in the heavier isotopes. In 106Ru and 108Ru the energy spread of the triplet would be even larger than the phonon energy itself and the centroid would occur at an energy more than three times that of the first 2+ state. The nature of these 0+ states is, however, still an open question. An explanation via a second minimum in the potential energy surface seems to fail in these cases. The rather strong excitation of the excited 0+ states in the 102,104Ru(t,p)104,106Ru reactions with 20% and 15%, respectively, of the ground state strength, leads to the assumption that an influence of pairing vibrations might be taken into consideration for these nuclei.

Only a smooth decrease in the energy of the first excited 2+ state with increasing neutron number is observed for the even palladium isotopes.
(Fig. 6). The $E_{2+}/E_{4+}$ ratio keeps almost constant at a value of about 2.4. The low-lying second excited $2^+$ states, which are even below the $4^+$ states, indicate the extreme softness of these nuclei in the $\gamma$ direction, and are responsible for the destruction of the rotational structure. The $B(E2)$ ratios of the transitions from the second $2^+$ state to the first $2^+$ state and to the ground state are well described by the Davydov model\(^1\). In the calculations of Faessler et al.\(^4\) flat minima at $\gamma = 300^\circ$ resulted for $106,106,110^{0}\text{PD}$ with a deformation parameter of $\beta = 0.17$.

For the odd-mass nuclei in this region the $\gamma$-ray spectra and decay schemes are rather complex and even the assignment of the ground state configuration is problematic in most cases. As an example the systematic trends of the low-lying states in odd Ru isotopes are shown in Fig. 7. The ground state of $^{99,101}\text{Ru}$ can be identified with a $d_{5/2}$ single particle state in agreement with the shell model. The energy of the $(J = 1)$ state decreases in $^{100}\text{Ru}$ and $^{102}\text{Ru}$ and shifts 2.9 and 20.6 keV, respectively, below the $5/2^+$ state, representing now the ground state. An explanation for this behaviour has been given by Rekstad\(^6\) in the framework of the Nilsson model with strong Coriolis coupling.

To summarize: the decay properties of neutron-rich nuclei in the mass region $A > 100$ indicate extreme softness, in agreement with recent theoretical calculations. They represent an interesting class of nuclei which should be investigated in more detail, at least in some representative cases, by extensive $\gamma$-$\gamma$ coincidence and angular correlation measurements. This requires, however, on-line separation methods. We have started such measurements at the fission product separator 'LOHENGRIN' in collaboration with a group from Grenoble. Furthermore, we are going to apply the gas jet recoil technique combined with the fast chemical on-line separation system 'SISAR'.

References

5. N. Trautmann, these proceedings.
8. Nuclear Data Sheets, ed. by Nuclear Data Group (Oak Ridge National Laboratory, Oak Ridge, Tennessee).