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Synthesis and Reanalysis of Light Short-lived Actinides

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Short-lived α emitters with N = 128-131 and Z = 91-93 were studied in the fusion reaction ^{40}Ar + ^{187}Re using the digital pulse processing technique at the gas-filled recoil separator Spectrometer for Heavy Atom and Nuclear Structure (SHANS). Two new isotopes ^{223,224}Np were identified and the decay of 220Pa was re-investigated through temporal and spatial correlations. The pileup signals were resolved by using the digital pulse processing technique. An α decay with half-life of $T_{1/2} = 2.15(^{100}_{52})$ μs and energy of E_α = 9477(44) keV was attributed to ^{223}Np. Two α-decay branches with half-life of $T_{1/2} = 38(^{26}_{11})$ μs, α energies of 9137(21) keV and 8868(60) keV were assigned to ^{224}Np, decaying to two excited states in ^{220}Pa. The results of ^{223}Np disprove the existence of a Z = 92 subshell closure.

KEYWORDS: new isotopes, digital pulse processing technique, super-pulse algorithm, subshell closure

1. Introduction

The evolution of proton shell structure beyond ^{208}Pb is of decisive importance for the shell stabilization of superheavy elements. The existence of a subshell or even shell gap at Z = 92 between the proton $h_{9/2}$ and $f_{7/2}$ orbitals has been a topic of intense theoretical debate. A substantial Z = 92 shell gap is predicted in many relativistic mean-field calculations like an early work for heavy elements [1], in most of the covariant density functionals (CDFs) [2,3] and also in some non-relativistic models [4]. Macroscopic-microscopic calculations [5] predicted a subshell gap at $Z = 92$. This is at variance with large-scale shell-model calculations [6], which show no sign of a shell gap at $Z = 92$ for $N = 126$ isotones and are in overall agreement with spectroscopic data on these isotones up to U [6–10].

The proton separation energy, ground-state spin and parity of odd-Z isotopes beyond U, e.g. Np isotopes ($Z = 93$), could help clarify the absence/presence of the $Z = 92$ subshell closure. Such experimental verification is necessary and valuable for testing the nuclear structure models and understanding the nature of nuclear forces as well [11–13]. However, for isotopes of elements above lead and far off the β-stability line, α decay prevails as the major radioactive decay mode and α spectroscopy is an indispensable tool to investigate the low-energy structure of heavy neutron-deficient nuclei. The spin and parity of Np isotopes can be tentatively proposed by the preformation probability of α particle.

All over the chart of nuclides, the region to the "north-east" of ^{208}Pb, with $Z \geq 84$ and $N = 128$-130, hosts the shortest-lived α radioactivities, with half-lives in the range of nanoseconds to microseconds. So far ^{219}Pa ($T_{1/2} = 53$ ns) [14] with $N = 128$ is among the shortest-lived α emitters, another example is ^{104}Te [15], just above the heaviest self-conjugate doubly magic ^{100}Sn. Synthesis and detection of neutron-deficient isotopes above thorium in this region are challenging due to their low production cross sections and short half-lives. With increasing atomic number Z, the fission probability of the compound nucleus increases rapidly and the evaporation of protons and α particles is by far dominant over neutron evaporation [16]. Progress in this region has been very slow in the last three decades, the frontier in this region was only pushed forwards from Pa to U [17].

In this proceeding, we report on the first observation of the short-lived neptunium isotopes ^{223,224}Np. In this experiment some fusion evaporation residues (ER) products and decay products have neutron number $N = 128$-130 and are very short-lived and their singals pileup. With conventional analog electronics, the shortest half-lives accessible are around tens of μs, it is extremely difficult or impossible to resolve these pileup signals in either energy or time. In recent years, digital pulse processing has been successfully applied to resolve such pileup events in the charged particle spectroscopy of short-lived nuclei [17–19].

2. Experiment

The experiment of ^{40}Ar+^{187}Re reaction were performed at the gas-filled recoil separator SHANS. The ^{40}Ar beam was accelerated to 188 MeV by the Sector-Focusing Cyclotron (SFC) of the Heavy Ion Research Facility in Lanzhou (HIRFL). Details of the experiment setups were described in our published papers [20–22].

Energy calibrations were performed with ^{175}Lu, ^{186}W and ^{187}Re targets at the same beam energy, covering a range of 6-19 MeV, specifically 6.3-9.4 MeV for single α energy and up to 19 MeV for double α sum energy. For non-pileup traces of long-lived α radioactivities, a trapezoidal filter with rising time 5 μs and flat top 3 μs was used to extract the full pulse height [23]. The energy resolution (FWHM) obtained with all vertical (horizontal) strips summed up is 22 (30) keV at α-particle energy of $\sim$7000 keV.

In [20–22], for pileup events depending on the time difference ΔT between the overlapping signals, the energies of individual signals were extracted using different algorithms. For overlapping signals with $\Delta T = 0.5$-15 μs, a trapezoidal filter with rising time 200 ns and flat top 200 ns was used, and the energy resolution (FWHM) of single α-energy is about 55 keV. For signals with $\Delta T = 200$-500 ns, the pulse-height of individual signal was obtained from the difference between the average of about six data points in the plateau area after the leading edge and that before the leading edge (average difference algorithm).The energy resolution of vertical strips for α decays recorded in double/multiple pulse traces with ΔT

down to ∼0.5 μs and ∼0.2 μs are around 55 keV and 70 keV, respectively. In the interval $\Delta T = 100$-200 ns, the average difference algorithm was applied but with smaller number of data points, the energy resolution obtained is around 140 keV. This indicates that as ΔT gets shorter, the energy resolution gets worse dramatically.

Our Peking University collaborators X. Wang, et al., have recently developed an improved super-pulse algorithm for fitting pulse waveforms [24]. Using this method, The energy resolution can be improved significantly to ∼28 keV for pileup pulses with $\Delta T > 150$ ns. For even shorter time difference, $\Delta T < 100$ ns, the boundary between the two α pulses is difficult/impossible to determine. In such cases, the individual α energy may be extracted using the pulse height of each α, but the results will be rather arbitrary and unreliable.

3. Results

3.1 The New Isotope ^{223}Np

The $N = 129$ isotones ^{219}Th (ER) and ^{218}Ac (daughter of ^{222}Pa implant) have half-lives of around 1 μs, while the $N = 128$ isotone ^{216}Ra (daughter of ^{220}Th implant) has a half-life of 0.18 μs, making them suitable benchmarks for ER-α_1 or α_1-α_2 pileup trace analysis. The resolved α energy spectra and the decay curves for ^{219}Th, ^{218}Ac and ^{216}Ra are shown in [20]. The α energies and half-lives measured in the present work are in good agreement with the literature values [25].

In order to identify decay chains belonging to ^{223}Np, all digital traces correlated to the subsequent α decay of ^{215}Ac ($E_\alpha = 7600(4)$ keV, $T_{1/2} = 0.17(1)$ s) [25], which are the third members of the α-decay chain originating from ^{223}Np, were checked event by event for the presence of multiple pulses. Ten multiple traces, all of which are triple pulse traces ER-α_1-α_2, were unambiguously attributed to the implantation of ^{223}Np followed by α decays of ^{223}Np and ^{219}Pa [20].

The α energy of ^{223}Np, deduced as the difference between the sum energy and the α energy of ^{219}Pa, is 9477(44) keV. The half-life of ^{223}Np was determined to be $2.15(^{100}_{52})$ μs by averaging the time differences between ^{223}Np implantations and decays, the errors were calculated following the method in Ref. [26].

Spin and parity of $9/2^-$ were tentatively proposed for the ground state of ^{223}Np by combining the reduced α-decay width and large-scale shell-model calculations. This assignment together with the proton separation energy disprove the existence of a $Z = 92$ subshell closure. Details of this discussion can be found in published paper about ^{223}Np [20].

3.2 The New Isotope ^{224}Np

Similarly, new isotope ^{224}Np can be identified by establishing its α decay sequences, terminated with the α-decay of ^{220}Pa or ^{216}Ac [21, 27], as ^{212}Fr ($b_\alpha \approx 43\%$), the daughter of ^{216}Ac, has a half-life of 20(6) minutes [25], too long to be correlated in this experiment. Six correlated decay chains were assigned to ^{224}Np [22].

In these decay chains, the α-decay signals of ^{224}Np and ^{220}Pa were piled up in the 15 μs recording cycle due to the short decay times of ^{220}Pa. The sum of the Q_α values for the two decay paths proposed above are 19106(30) keV and 19094(86) keV, respectively, identical within the error bars, implying that the two decay paths start from one same level in ^{224}Np. A half-life of $T_{1/2} = 38(^{26}_{11})$ μs and α energies of 9137(21) keV and 8868(60) keV were determined for the two α branches in ^{224}Np. The more detailed discussion and the decay scheme proposed are shown in published paper [22].

3.3 Reinvestigation of the Short-lived α-emitter ^{220}Pa

The short-lived implanted ERs were identified based on the parent-daughter decay correlations. In the most intense activity, a half-life of 0.36(7)ms and E_α =9.069(3) MeV were deduced for α decay of the daughter nucleus, in good agreement with the literature value of ^{216}Ac [25]. Therefore the parent activity was unambiguously identified as 220Pa. Then an α energy of E_α =9.520(16) MeV and a half-life of $T_{1/2}$ =0.90(13)s were deduced for ^{220}Pa [21].

Spin and parity of 1^- were tentatively assigned to the ground state of ^{220}Pa based on the systematics of the reduced α-decay widths of the N =129 isotones. Detailed information of this analysis can be found in published paper about ^{220}Pa [21].

3.4 Re-analysis of the data for ^{223,224}Np Using Improved Super-pulse Algorithm

Super-pulse algorithm is a method for fitting digital pulse waveforms. The details of this method are described in [28]. On this basis, X. Wang, et al. of Peking University further improved the algorithm and called it improved super-pulse algorithm. Specific improvements include baseline correction of waveforms and optimization of algorithm for fitting pileup waveforms. The details of improved super-pulse algorithm are introduced in [24].

The data of ^{223,224}Np was re-analyzed by using improved super-pulse algorithm. For ^{223}Np, in addition to the ten events reported in [20], three new events were found. The decay chains corresponding to three new events are listed in Table I. Combining the information of these 13 events, a new half-life of $T_{1/2} = 2.63(^{101}_{57})$ μs and energy of $E_\alpha = 9499(36)$ keV were determined for ^{223}Np. Compared with the $T_{1/2} = 2.15(^{100}_{52})$ μs and $E_\alpha = 9477(44)$ keV reported in [20], they are in agreement with each other and the new results has improved precision in energy.

Table I. Three new decay chains attributed to ^{223}Np. α_i represents α particle from ^{223}Np, ^{219}Pa and ^{215}Ac, for i = 1, 2 and 3, respectively. The units are keV for the implantation energy of ER and α particle energies. The column $(E_{\alpha1} + E_{\alpha2})$ lists the sum energy of two overlapping α signals.

Event No.	E_{ER}	$E_{\alpha1}$	$E_{\alpha2}$	$(E_{\alpha1} + E_{\alpha2})$	$E_{\alpha3}$	$T_{\alpha1}/\mu$s	$T_{\alpha2}$/ns	$T_{\alpha3}$/ms
1	12502	9462$^{a)}$	10010$^{a)}$	19472	1488	15.75	40	200.13
2	11257	9392$^{a)}$	697$^{a)}$	10089	7627	2.00	60	91.1
3	14587	412	9972	10386	944	0.82	100	72.2

a) The α energies extracted from α(^{223}Np)-α(^{219}Pa) pileup pulses with time differences shorter than 100 ns cannot be reliably quantified.

Similarly, one new event of ^{224}Np was found by using this method and the results for ^{224}Np were also improved, with $T_{1/2} = 49(^{40}_{17})$ μs and α energies of 9142(18) keV and 8868(37) keV, in agreement the results reported in [22].

4. Summary

In summary, new short-lived isotope ^{223,224}Np were synthesized in the fusion reaction ^{40}Ar + ^{187}Re and identified. The half-lives and α-decay energy were extracted from pileup pulse waveforms by using two different algorithms, i.e., average difference and improved super-pulse algorithm. The results show that the super-pulse algorithm is more suitable for analyzing pileup waveforms. The results of ^{223}Np support the non-existence of the Z = 92 subshell closure.

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