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## Production and decay of the heaviest odd-Z nuclei in the $^{249}\text{Bk} + ^{48}\text{Ca}$ reaction

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**Abstract.** The reaction of  $^{249}\text{Bk}$  with  $^{48}\text{Ca}$  has been investigated with an aim of synthesizing and studying the decay properties of isotopes of the new element 117. The experiments were performed at five projectile energies (in two runs, in 2009-2010 and 2012) and with a total beam dose of  $^{48}\text{Ca}$  ions of about  $9 \times 10^{19}$ . The experiments yielded data on  $\alpha$ -decay characteristics and excitation functions of the produced nuclei that establish these to be  $^{293}117$  and  $^{294}117$  – the products of the  $4n$ - and  $3n$ -evaporation channels, respectively. In total, we have observed 20 decay chains of  $Z=117$  nuclides. The cross sections were measured to be 1.1 pb for the  $3n$  and 2.4 pb for the  $4n$ -reaction channel. The new  $^{289}115$  events, populated by  $\alpha$  decay of  $^{293}117$ , demonstrate the same decay properties as those observed for  $^{289}115$  produced in the  $^{243}\text{Am}(^{48}\text{Ca}, 2n)$  reaction thus providing cross-bombardment evidence. In addition, a single decay of  $^{294}118$  was observed from the reaction with  $^{249}\text{Cf}$  – a result of the in-growth of  $^{249}\text{Cf}$  in the  $^{249}\text{Bk}$  target. The observed decay chain of  $^{294}118$  is in good agreement with decay properties obtained in 2002-2005 in the experiments with the reaction  $^{249}\text{Cf}(^{48}\text{Ca}, 3n)^{294}118$ . The energies and half-lives of the odd-Z isotopes observed in the 117 decay chains together with the results obtained for lower-Z superheavy nuclei demonstrate enhancement of nuclear stability with increasing neutron number towards the predicted new magic number  $N=184$ .

### 1. Introduction

After many unsuccessful attempts to reach the predicted “island of stability” of the superheavy elements (SHE) by means of different methods (see, e.g., Review [1] and references therein), including usage of kinematic separators of the fusion-evaporation reaction products [2], the work aimed at achieving this goal has been resumed in the Flerov Laboratory of Nuclear Reactions (JINR, Dubna).

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For the first time, in complete-fusion reactions  $^{242,244}\text{Pu}$ ,  $^{245,248}\text{Cm}$ , and  $^{249}\text{Cf}$  targets with  $^{48}\text{Ca}$  projectiles we have synthesized nine isotopes of even- $Z$  nuclei of the new elements 114 (Fl), 116 (Lv) and 118 employing the Dubna gas-filled recoil separator (DGFRS) [3]. The production cross-sections and decay properties of the isotopes of  $^{286-289}\text{114}$  and  $^{292,293}\text{116}$  were recently confirmed in independent experiments performed at FLNR [4], LBNL [5,6], and GSI [7-9].

The next step towards the island of stability of the nuclei around  $N=184$  and  $Z=114$  (or 120-126) was the production of odd- $Z$  nuclides in reactions with odd- $Z$  targets ( $^{237}\text{Np}$ ,  $^{243}\text{Am}$ , and  $^{249}\text{Bk}$ ). Studies of odd- $Z$  nuclei provide even more detailed information about the structure of the heaviest nuclides than studies of even- $Z$  nuclei because of the longer odd- $Z$  decay chains; this results from strong fission hindrance caused by the unpaired nucleons. For the first time, the nuclei  $^{287}\text{115}$  and  $^{288}\text{115}$  and their decay products including  $^{283}\text{113}$  and  $^{284}\text{113}$  isotopes were observed in 2003 [10] in the  $^{243}\text{Am}+^{48}\text{Ca}$  reaction; then new isotopes  $^{289}\text{115}$  and  $^{285}\text{113}$  were observed in 2010-2012 [11]. In 2006 a lighter isotope  $^{282}\text{113}$  was synthesized in the  $^{237}\text{Np}(^{48}\text{Ca},3n)$  reaction [12]. In addition, three decay chains of the lightest isotope  $^{278}\text{113}$  were observed in the reaction  $^{209}\text{Bi}(^{70}\text{Zn},n)$  in 2004, 2005, and 2012 [13].

The discovery of element 117 in the  $^{249}\text{Bk}+^{48}\text{Ca}$  reaction has been reported in 2010 [14]. A relatively high stability of all these odd- $Z$  nuclei is caused by the influence of presumably spherical nuclear shells at  $Z=114-126$  and  $N=184$ . In 2012, we have performed a new series of experiments to obtain a more detailed information on the decay properties of odd- $Z$  nuclei, to measure the excitation function of the  $^{249}\text{Bk}+^{48}\text{Ca}$  reaction in a more extended range of projectile energies, and to make a cross-bombardment consistency check of the reported discoveries of elements 113, 115 and 117 [11,15].

## 2. Experiment

For the synthesis and identification of these odd- $Z$  nuclei, we used the Dubna gas-filled recoil separator that selects evaporation products of complete-fusion reactions that are strongly forward peaked and suppresses the products of transfer reactions and reactions with emission of charged particles ( $pxn$ ,  $\alpha xn$ , etc.). The evaporation residues were separated in flight, then passed through a time-of-flight system and were implanted in a focal-plane silicon detector, with an estimated transmission efficiency of about 35% for  $Z=117$  nuclei. Prior to 2011 in our work we used  $4\times 12\text{ cm}^2$  Si-detector array with 12 vertical position-sensitive strips surrounded by eight  $4\times 4\text{ cm}^2$  detectors without position sensitivity (see Refs. [3,10-12,14], and references therein). The detection system was calibrated by registering the recoil nuclei and decays ( $\alpha$  or SF) of the known isotopes of No and Th and their descendants [16] produced in the reactions  $^{206}\text{Pb}(^{48}\text{Ca},2n)$  and  $^{\text{nat}}\text{Yb}(^{48}\text{Ca},3-5n)$ , respectively. In the above experiments, the full-width-at-half-maximum (FWHM) energy resolution of  $\alpha$  particles implanted in the focal-plane detectors was 60–140 keV, depending on the strip and the position within the strip. The FWHM position resolutions of the signals of correlated decays of nuclei implanted in the detectors were 1.1–1.3 mm for ER- $\alpha$  signals and 0.4–0.8 mm for ER-SF signals [14].

In 2011, before performing the new run for additional study of the excitation function of the  $^{249}\text{Bk}+^{48}\text{Ca}$  reaction and decay properties of  $Z=117$  isotopes and descendant nuclei, the detection system was modified to increase the position granularity of the detectors, which reduces the probability of observing sequences of random events that mimic decay chains of synthesized nuclei. The new focal-plane detectors consisted of two  $6\times 6\text{ cm}^2$  detectors each having 16 strips surrounded by six  $6\times 6\text{ cm}^2$  side detectors. The appropriate multi-channel data acquisition system [17] was developed by the DGFRS research crew on the basis of POLON spectroscopic modules and products of TekhInvest Ltd. (Dubna, Russia). This allowed us to get for the focal-plane detector the FWHM energy resolution of 34–73 keV, while the sum signals recorded by the side and focal-plane detectors had an energy resolution of about 83 to 117 keV. The FWHM position resolutions of the implantation detector were 1.1–1.8 mm for ER- $\alpha$  signals and 0.5–1.2 mm for ER-SF signals [15]. Other experimental conditions were the same as in Refs. [3,10-12,14]. The summary conditions of the  $^{249}\text{Bk}+^{48}\text{Ca}$  reaction studied at the DGFRS are shown in Table 1. In order to reduce the background rate in the detector, the beam was switched off (like it was implemented in all the previous

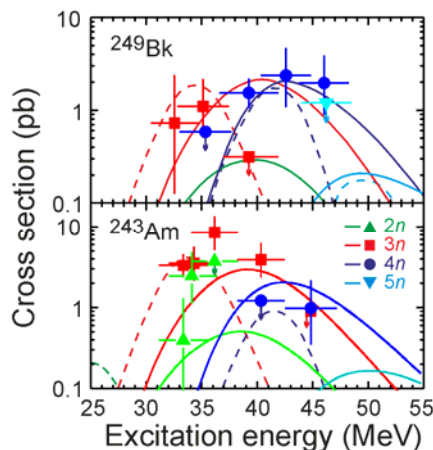
experiments) after a recoil signal was recorded being followed by an  $\alpha$ -like signal in the focal-plane detector within energy and time intervals corresponding to decays of parent and/or daughter nuclei, in the same strip and in close position.

**Table 1.** The  $^{249}\text{Bk}$  target thickness <sup>a</sup>, lab-frame beam energies in the middle of the target layers, resulting excitation-energy intervals, total beam doses, and numbers of observed decay chains assigned to the parent nuclei  $^{293}117$  ( $4n$ ) and  $^{294}117$  ( $3n$ ) are listed.

Target thickness (mg/cm <sup>2</sup> )	$E_{\text{lab}}$ (MeV)	$E_{\text{exc}}$ (MeV)	Beam dose $\times 10^{18}$	Number of chains $4n / 3n$	Ref.
0.33	243.7	30.4–34.7	9.4	0 / 1	[15]
0.31	247.0	33.2–37.5	20.	0 / 1	[14]
0.33	246.8	32.8–37.5	5.4	0 / 2	[15]
0.31	251.7	37.2–41.4	24.	5 / 0	[14]
0.33	251.7	37.0–41.9	14.1	5 / 0	[15]
0.33	255.7	40.3–44.8	9.2	3 / 0	[15]
0.33	259.8	43.8–48.3	11.9	3 / 0	[15]

### 3. Results

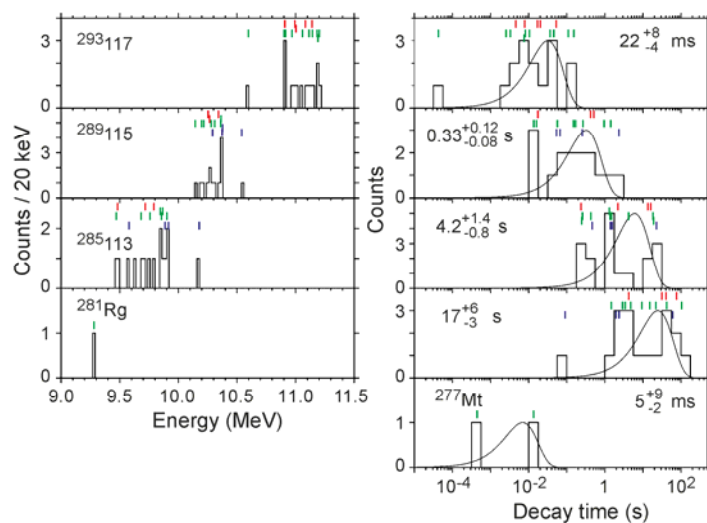
In the experiments with the  $^{243}\text{Am}+^{48}\text{Ca}$  reaction in 2010–2012, at the lowest excitation energies  $E^*=31.1$ – $36.4$  MeV four decay chains of the isotope  $^{289}115$ , the product of the  $2n$  channel, were observed [11] (see Figure 1). Because of the mass difference between  $^{243}\text{Am}$  and  $^{249}\text{Bk}$  ( $\alpha+2n$ ) and lower yields of the  $1n$  and  $5n$  channels compared with the  $2$ - $4n$  channels, one and the same isotope of element 115,  $^{289}115$ , can be produced only in the  $2n$ - and  $4n$ -evaporation channels of the  $^{243}\text{Am}+^{48}\text{Ca}$  and  $^{249}\text{Bk}+^{48}\text{Ca}$  reactions, respectively. Indeed, 16 decay chains of the parent isotope  $^{293}117$  were observed in the  $^{249}\text{Bk}+^{48}\text{Ca}$  reaction at higher excitation energies  $E^*=37.0$ – $48.3$  MeV. The radioactive decay properties of  $^{293}117$  and all the descendant nuclei discovered in 2010 [14] were confirmed by registration of 11 new decay chains in this new series of experiments [15]. One can see in Figure 2 that decay characteristics of  $^{289}115$ ,  $^{285}113$ , and  $^{281}\text{Rg}$ ,  $\alpha$ -decay products of  $^{293}117$ , in the five events observed in the first experiment [14] and four events originating from  $^{289}115$  and produced in cross reaction with  $^{243}\text{Am}$  [11] are in good agreement with the recent data. Thus, the isotope  $^{289}115$  was produced in two reactions with target nuclei  $^{243}\text{Am}$  and  $^{249}\text{Bk}$  that provide cross-bombardment evidence for the discovery of elements 117, 115, and 113.



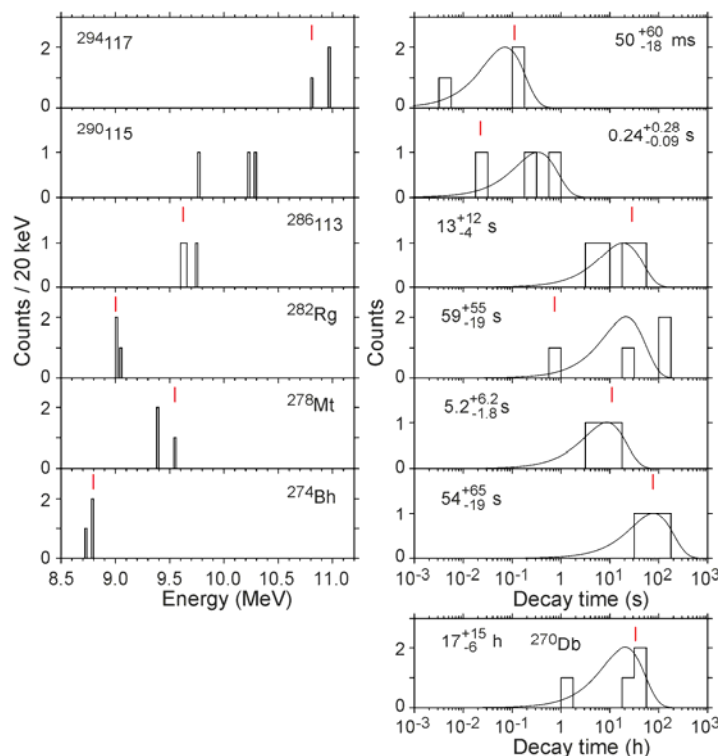
**Figure 1.** Measured cross sections of the  $xn$ -evaporation channels of the  $^{249}\text{Bk}+^{48}\text{Ca}$  and  $^{243}\text{Am}+^{48}\text{Ca}$  reactions. Vertical error bars correspond to statistical uncertainties [18]. Horizontal error bars represent the range of excitation energies populated at given beam energy. Symbols with arrows show upper cross-section limits. The results of theoretical calculations are shown by solid [19] and dashed [20] lines.

The heaviest isotope  $^{294}117$ , product of the  $^{249}\text{Bk}(^{48}\text{Ca},3n)$  reaction, was synthesized at lower excitation energies of 30.4–37.5 MeV. The decay properties of all the nuclei determined in the four decay chains originating from parent nucleus  $^{294}117$  are shown in Figure 3. The properties of the nuclei in the new decay chains point to the same activities

<sup>a</sup> As in [14], the  $^{249}\text{Bk}$  was produced at ORNL at the High Flux Isotope Reactor. The Bk fraction was chemically separated and purified at the Radiochemical Engineering Development Center at ORNL. Six arc-shaped targets were made at RIAR.



**Figure 2.**  $\alpha$ -particle energy spectra registered by the focal-plane detector only or together with the side one (left-hand panel) and decay-time distributions on a logarithmic scale (right-hand panel) for isotopes originating from  $^{293}\text{117}$ . The events originating from  $^{293}\text{117}$  observed in the first [14] and in the second experiment with  $^{249}\text{Bk}$  [15] and in the reaction with  $^{243}\text{Am}$  [11] are shown in the histograms by red (top), green (middle), and blue (bottom) lines, respectively.



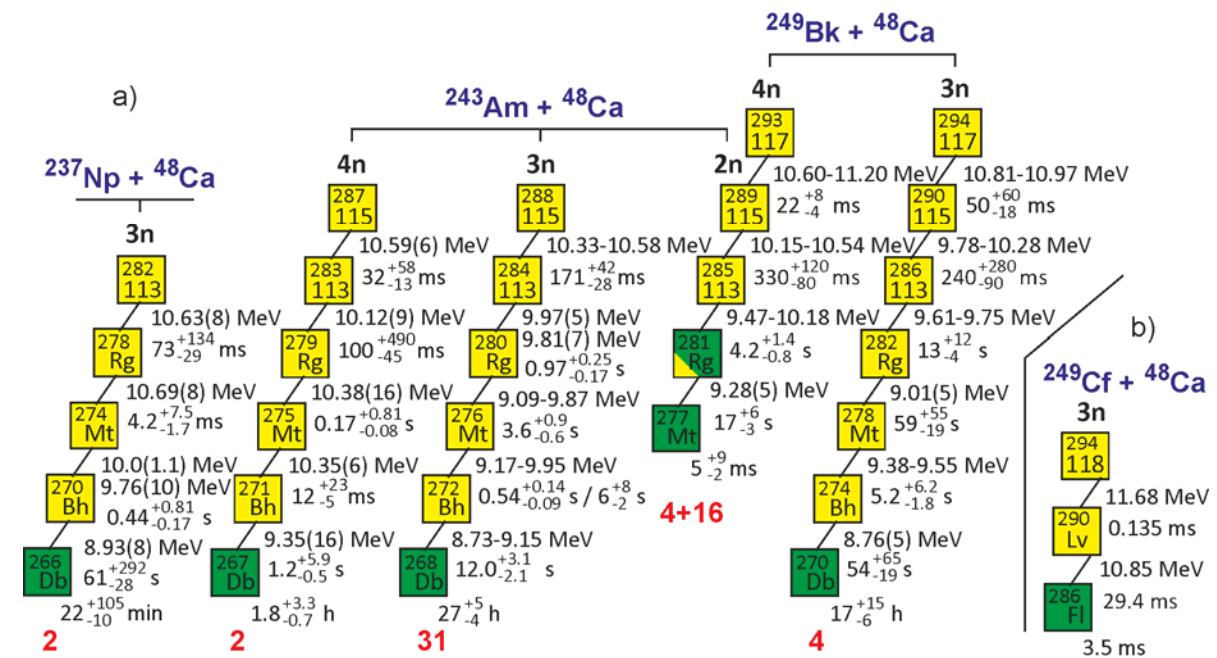
**Figure 3.** The same as Figure 1 but for  $^{294}\text{117}$ . The vertical lines show the energies and decay times of the events obtained in the first experiment [14].

arising from  $^{294}\text{117}$  detected in the two experiments using the  $^{249}\text{Bk}$  target [14,15].

The maximum cross sections for the  $3n$  and  $4n$  evaporation channels, were measured to be  $\sigma_{3n}=1.1^{+1.2}_{-0.6}$  pb and  $\sigma_{4n}=2.4^{+3.3}_{-1.4}$  pb, at  $E^*=35$  and  $43$  MeV respectively (see Figure 1).

The target isotope  $^{249}\text{Bk}$  with a half-life of 330 d decays into  $^{249}\text{Cf}$ . During a long experiment, this creates an opportunity to produce  $Z=118$  isotopes in the  $^{249}\text{Cf}+^{48}\text{Ca}$  reaction [21]. Indeed, with 247-MeV  $^{48}\text{Ca}$  we observed one more decay chain of nuclei whose radioactive properties agree well with those determined for  $^{294}\text{118}$  and its descendant nuclei  $^{290}\text{Lv}$  and  $^{286}\text{Fl}$  (fission branch of about 50%) [3,21] (see Figure 4b). Taking into account the buildup of  $^{249}\text{Cf}$  in the preceding [14] and present [15] experiments, the detected decay chain of  $^{294}\text{118}$  corresponds to  $0.3^{+0.7}_{-0.26}$  pb for the total excitation energy interval of 26.6–37.5 MeV of the compound nucleus  $^{297}\text{118}$ , in good agreement with cross sections already measured in this reaction [3,21].

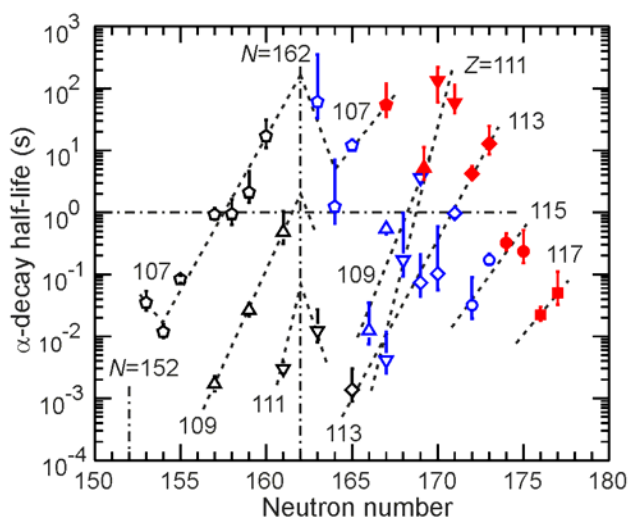
Therefore, the discovery of the chemical elements with atomic numbers 113, 115, and 117 that were synthesized for the first time in 2003 [10] and 2010 [14] has now been corroborated through the observation of additional decay chains in the reaction  $^{249}\text{Bk}+^{48}\text{Ca}$ . In total, 59 decay chains originating from odd- $Z$  parent nuclei  $^{282}\text{113}$  [12],  $^{287-289}\text{115}$  [10,11], and  $^{293,294}\text{117}$  [14,15] were observed and radioactive decay properties of 29 new isotopes were determined (see Figure 4a). The measured  $\alpha$ -particle energies for all the isotopes of odd- $Z$  elements produced in the reactions  $^{243}\text{Am}+^{48}\text{Ca}$  and  $^{249}\text{Bk}+^{48}\text{Ca}$  agree well with the systematics of the  $\alpha$ -decay energies of the heavy nuclei and have intermediate values between



**Figure 4.** (a) Summary of the decay properties of the isotopes of elements 113, 115 and 117 observed for the first time among the reaction products of  $^{48}\text{Ca}$  beam with  $^{237}\text{Np}$ ,  $^{243}\text{Am}$  and  $^{249}\text{Bk}$  targets. The number of the detected decay events of a given isotope is shown at the bottom of the chains. (b) Decay properties of  $^{294}\text{118}$ – $^{286}\text{119}$  measured in the  $^{249}\text{Cf}(^{48}\text{Ca}, 3n)$  reaction [14].

neighboring even- $Z$  nuclei. The measured  $\alpha$ -particle energies of the  $Z=107$  and  $Z=109$  isotopes as well as their behavior vs. neutron number are in full agreement with what is observed for the previously known neighboring lighter nuclei. Moreover,  $^{268}\text{Db}$ , the descendant nucleus of  $^{288}\text{115}$  and  $^{284}\text{113}$ , was identified in independent chemistry experiments [22,23].

The experimental half-lives of the isotopes of odd- $Z$  elements are shown in Figure 5. The increase of neutron number in nuclei with  $N \geq 165$  results in considerable rise of their half-lives. An especially strong growth of lifetimes with increasing  $N$  is observed for the isotopes of elements 109, 111, and 113. The half-lives, as well as  $\alpha$ -particle energies of the odd- $Z$  isotopes observed in the  $Z=117$  decay chains, together with the results obtained for lower- $Z$  superheavy nuclei, demonstrate the decisive role



**Figure 5.** Half-lives vs. neutron number for the isotopes of odd- $Z$  elements (results from  $^{249}\text{Bk} + ^{48}\text{Ca}$  reactions are shown by full red diamonds).

of nuclear shell effects resulting in enhancement of stability with increasing neutron number towards the predicted new magic number  $N=184$ .

Experimental study of the properties of odd- $Z$  nuclei was recently performed at the gas-filled recoil separator TASCA at GSI in fusion-evaporation reactions  $^{243}\text{Am} + ^{48}\text{Ca}$  [24] and  $^{249}\text{Bk} + ^{48}\text{Ca}$  [25]. Authors of Ref. [24] announced observation of 23 correlated  $\alpha$ -decay chains starting from  $^{287,288}\text{115}$  isotopes; their observed decay pattern and properties are in good agreement with the DGFRS data [10,11]. Two decay chains of  $^{294}\text{117}$  and descendant nuclei observed in the  $^{249}\text{Bk} + ^{48}\text{Ca}$  reaction have been reported in [25]. The production cross



section of  $^{294}_{117}$  nucleus and decay properties of the observed descendant nuclei confirm previously reported DGFRS data [14,15].

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