Attempt to produce element 120 in the ^{244}Pu + ^{58}Fe reaction

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An experiment aimed at the synthesis of isotopes of element 120 has been performed using the ^{244}Pu(^{58}Fe,xn)$^{302-x}$120 reaction. No decay chains consistent with fusion-evaporation reaction products were observed during an irradiation with a beam dose of 7.1×10^{18} 330-MeV ^{58}Fe projectiles. The sensitivity of the experiment corresponds to a cross section of 0.4 pb for the detection of one decay.

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I. INTRODUCTION

During recent years, significant progress has been achieved in the experimental and theoretical investigations of the region of superheavy nuclei. The macro-microscopic nuclear model (MM) predicts a substantial enhancement of the stability of heavy nuclei when approaching the closed spherical shells at $Z = 114$ and $N = 184$ [1]. Later microscopic Hartree-Fock-Bogoliubov (HFB) and relativistic mean field (RMF) theories predict a large stabilizing effect of the neutron shell at $N = 184$ but also suggest that the magic proton shell should be at higher proton numbers $Z = 120$–126 [2].

In our previous experiments, 18 new nuclides with $Z = 112$–118 and their daughter isotopes have been produced in complete fusion reactions of actinide target nuclei (^{238}U–^{249}Cf) with ^{48}Ca beams [3]. The decay properties of these nuclei revealed a significant increase in their stability as they approached the predicted neutron shell $N = 184$. The nuclides with the largest neutron and proton numbers that were synthesized in reactions with the heaviest target nuclei ^{248}Cm and ^{249}Cf [^{293}116 ($N = 177$) and ^{294}118 ($N = 176$), respectively] still possess 7–8 fewer neutrons than the predicted magic number $N = 184$. One can expect that increasing the number of neutrons in these nuclei would result in a further increase of their stability against decay. Unfortunately, nuclides with $Z > 98$ do not exist in quantities sufficient for constructing targets for these types of experiments. Therefore, isotopes of elements 116 and 118 with more neutron excess can be reached only if they are formed as α-decay products of the heavier evaporation residues. For this purpose, we need to use complete fusion reactions with projectiles heavier than ^{48}Ca. One should note also that increasing the atomic number brings us closer to the predicted closed proton shell at $Z = 120$–124, which should also increase shell effects.

Three reactions, ^{238}U + ^{64}Ni, ^{244}Pu + ^{58}Fe, or ^{248}Cm + ^{54}Cr, can be used for synthesis of isotopes of element 120, all leading to the same compound nucleus, ^{302}120 ($N = 182$). In the present experiment, the fusion of the nuclei ^{244}Pu and ^{58}Fe was used.

In the reaction ^{244}Pu + ^{58}Fe, the excitation energy at the Coulomb barrier ($B_{\mathrm{Bass}} = 310.5$ MeV) is $E^* = 29.4$ MeV. By analogy with the fusion reactions of actinide targets with ^{48}Ca ions [3] the maximum yield of the evaporation residues is expected for the $3n$- and $4n$-evaporation channels that result in the formation of the isotopes ^{299}120 and ^{298}120, respectively. Their expected decay schemes are presented in Fig. 1.

According to theoretical predictions (see Table I) the even-odd nuclide ^{299}120 ($N = 179$) should undergo α decay with a decay energy in the range of $Q_\alpha = 11.5$–13.23 MeV and a half-life of $T_\alpha \sim 1$ μs -10 ms (not taking into account the hindrance due to the odd neutron number). According to the estimates given in Ref. [15], the α-decay energy and half-life are $Q_\alpha = 13.06$ MeV and $T_\alpha = 15$ μs. At the Dubna Gas-Filled Recoil Separator (DGFRS) [16,17] the time of flight of the recoil nuclei from the target to the separator's focal plane is about 1 μs; thus the whole decay chain of the isotope ^{299}120 should be registered by the focal-plane detectors. The half-life of the even-even nucleus ^{298}120 as seen in Table I also considerably exceeds 1 μs, except for the two estimates obtained in the version of the MM model in Refs. [6] and [7]. Note that the experimental values of the half-lives of the lighter even-even nuclides ^{286,288}114, ^{290,292}116, and ^{294}118 are considerably higher than those predicted in Refs. [6] and [7]. Therefore, the whole decay chain of ^{298}120 also should be registered in the focal-plane detectors with high probability. Both isotopes of the element 120 will preferentially undergo α decay ($T_\alpha < T_{\mathrm{SF}}$) according to calculations and measured Q_α and T_α values of the nuclides with $Z \leqslant 118$. After the two sequential α decays of the nucleus ^{299}120 (see Fig. 1) the known 18-ms isotope ^{291}116 is formed, which has already been produced and characterized in the reaction ^{245}Cm(^{48}Ca, $2n$)^{291}116 [18,19]. The product of the α decay of the neighboring even-even nucleus ^{298}120 is a known 0.9-ms isotope of element 118 that has been synthesized in the reaction ^{249}Cf(^{48}Ca, $3n$)^{294}118 [18,19].

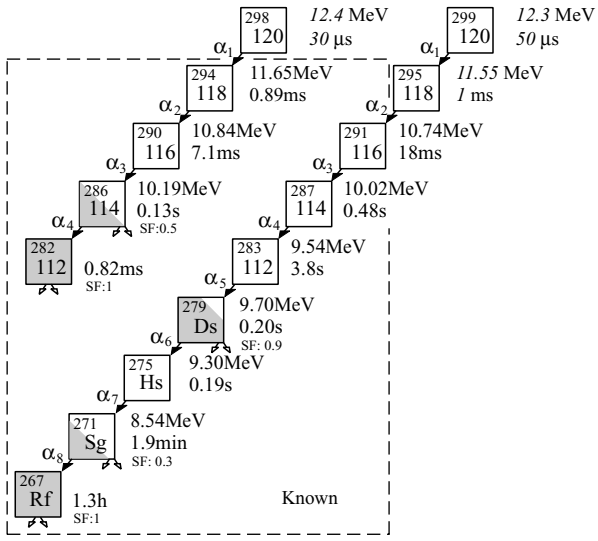

FIG. 1. Expected decay chains for $^{298,299}120$. The decay schemes for $^{294}118$ and $^{291}116$ are known from previous work, and the decay properties given in the figure are average experimental. The properties of $^{298,299}120$ and $^{295}118$ are calculated (see Table I and text).

II. EXPERIMENTAL DETAILS AND RESULTS

The experiment was performed in January–March of 2007 using the DGFRS apparatus. The ^{58}Fe-ion beam was accelerated by the U400 cyclotron. The typical beam intensity at the target was 0.7 pμA. A total beam dose of 7.1×10^{18} ^{58}Fe ions was accumulated.

The beam energy was determined by employing a time-of-flight system with a systematic uncertainty of about 1.4 MeV. The beam energy was also monitored by the observed yield of the isotopes $^{214-216}$Th produced in the $2n$- to $4n$-evaporation channels of the reaction $^{160}\mathrm{Gd} + {}^{58}\mathrm{Fe}$ at $E_{\mathrm{lab}} = 242.4$ MeV. For this reaction, a ^{160}Gd target was inserted before and periodically during the experiment, and the DGFRS settings were changed for Th.

The target consisted of the enriched isotope ^{244}Pu (98.6%, 0.37 ± 0.02 mg/cm^2) deposited as an oxide (PuO_2) onto 0.72 mg/cm^2 Ti foils. The beam energy in the middle of the target layer was chosen to be $E_{\mathrm{lab}} = 330.4$ MeV, which corresponded to an excitation energy of the compound nucleus of $E^* = 45.5$ MeV calculated using the masses of Refs. [20] and [21]. The range of excitation energies $\Delta E^* = \pm 2.2$ MeV takes into account the thickness of the target ($\Delta E^{\mathrm{tot}}_{\mathrm{lab}} = \pm 2.3$ MeV) and the energy spread of the incident cyclotron beam ($\Delta E_{\mathrm{lab}} = \pm 1.5$ MeV). The beam energy losses in the separator's entrance window and target backing and layer were calculated using the available data of Hubert, Bimbot, and Gauvin [22] or Northcliffe and Schilling in other cases [23].

The evaporation residues (ER) recoiling from the target were spatially separated in flight from beam particles, scattered particles, and transfer-reaction products and collected on the detectors in the focal plane of the separator with a transmission efficiency of $45 \pm 10\%$ for $Z = 120$ nuclei [16,17]. The average charge of heavy ERs in hydrogen (q) at a pressure of 1 Torr that is necessary for setting the separator was estimated to be $q = 8.5$ [24].

ER passed through a time-of-flight system and were implanted in a 4×12 cm semiconductor detector array with 12 vertical position-sensitive strips surrounded by eight 4×4 cm side detectors. The detection efficiency for full-energy α particles emitted in the decays of implanted nuclei was 87%. The FWHM energy resolutions were 55–95 keV (depending on the strip) for α particles absorbed in the focal-plane detector and 110–155 keV for α particles that escaped this detector with a low energy release and were subsequently registered by a side detector. The FWHM position resolutions of the signals of correlated decays of nuclei implanted in the detectors were 1.0–1.8 mm for ER-α signals and 0.4–0.9 mm for ER-spontaneous fission (ER-SF) signals.

For the detection of sequential decays of the daughter nuclides in the absence of a beam-associated background, the beam was switched off for 30 s after a recoil signal was detected with an implantation energy of $E_{\mathrm{ER}} = 6.5$–18.5 MeV, which is expected for complete-fusion evaporation residues (an additional degrader was placed before the detectors for reducing the ER energy so that it would have $E \leqslant 20$ MeV and be registered in the α-particle electronics), followed by one or more α-like signals with energies of $E_{\alpha 1} = 12$–12.7 MeV, $E_{\alpha 2} = 11.3$–11.9 MeV, $E_{\alpha 3} = 10.5$–11.1 MeV,

TABLE I. Calculated properties of element 120 isotopes, from the indicated sources. Half-lives corresponding to the predicted α-particle energies, calculated from the Viola-Seaborg systematics with parameters from Refs. [4] and [5], are shown in italics.

$^{298}120$			$^{299}120$			Model	Reference
Q_α (MeV)	T_α (ms)	T_{SF} (ms)	Q_α (MeV)	T_α (ms)	T_{SF} (ms)		
13.44	*0.0005*		13.23	*0.001*		MM	[6]
13.36	0.0004	30				MM	[7]
13.0	*0.003*	3				MM	[8]
11.6	10					HFB	[4,5]
12.8	*0.01*		11.5	*10*		HFB	[9]
10.9	*300*					HFB	[10]
12.7	*0.01*					HFB	[11]
11.1	*100*					HFB	[12,13]
11.2	*40*					RMF	[14]

and $E_{\alpha 4} = 9.8$–10.4 MeV, detected in the same strip, within a 2.8-mm-wide position window, and in time intervals of 0.5 ms, 15 ms, 150 ms, and 1 s, respectively. These energies and times correspond to those expected for the isotopes of elements 120, 118, 116, and 114 in the decay chains shown in Fig. 1. If during the first 30-s beam-off time interval an α particle with $E_\alpha = 9.3$–11.9 MeV was registered in any position of the same strip, the beam-off interval was automatically extended to 10 min. In the 10-min period, if other α particles with energies expected for heavy nuclei were observed, we could prolong the beam-off pause even more.

As was mentioned above, at an excitation energy of 40–45 MeV for the compound nucleus $^{302}120$, the yield of the $3n$- or $4n$-evaporation products is expected to have the largest probability. Decay chains that could be assigned to the isotopes of element 120 or its daughter nuclei were not observed in our experiment, neither within the beam-off nor the beam-on time intervals. If the α decay of a parent nucleus had occurred during the dead time of the electronic system ($t_{\rm dt} = 7$ μs for ER-α signals) or in flight through the separator, we would still find the decay chains of daughter nuclei (in the latter case, the sensitivity of the experiment would be lower by several times because of a reduced transmission efficiency for such nuclei). We also did not find short ER-SF correlated events ($t_{\rm dt} \geqslant 85$ μs) that could originate directly from the spontaneous fission of $^{298}120$ if the results of the calculations resulting in Table I were in error.

The sensitivity of the experiment corresponds to 0.4 pb for the detection of one event. The 84% upper cross-section limit was set at 0.7 pb by only taking statistical uncertainties into account and 1.1 pb by adding other experimental uncertainties.

III. DISCUSSION AND CONCLUSIONS

Let us compare the two reactions ^{244}Pu + ^{48}Ca [18,19] and ^{244}Pu + ^{58}Fe, which involve the same target but projectiles of different masses. In these reactions the compound nuclei $^{292}114$ and $^{302}120$, respectively, are formed with the same excitation energy $E^* \approx 40$–45 MeV that corresponds to the maximum yield of the evaporation residues mostly from the $3n$- and $4n$-evaporation channels. At the same time, the production cross section of the evaporation residues in the reaction ^{244}Pu + ^{58}Fe appears to be lower by more than an order of magnitude compared with that in the reaction ^{244}Pu + ^{48}Ca [18,19].

In fusion reactions of very heavy nuclei, the cross section for the production of the evaporation residues is determined in two sequential stages—the collective motion of the composite system that leads to the formation of the excited compound nucleus and the survivability of the compound nucleus in its transition to the ground state via neutron emission. Given that the probabilities of forming the compound nuclei $^{292}114$ and $^{302}120$ with the same excitation energy $E^* \approx 40$–45 MeV are equal, the observed difference in the experimental cross sections can be explained by higher fissionability of $^{302}120$. This is associated with the decrease of the amplitude of the shell correction and, as a consequence, with the decrease of the height of the fission barrier of the nuclei when moving off the shell $Z = 114$. A similar scenario is expected in the framework of the MM model. By contrast, in the HFB model with different sets of forces and in RMF calculations [25] the fission barrier height strongly increases on the approach to the shell $Z = 120$ (or possibly 122). In this case, the survivability of the heavy compound nucleus $^{302}120$ is higher than that of $^{292}114$. Therefore, one might expect, with the same assumption of equal compound nucleus formation probabilities, a higher yield of $Z = 120$ evaporation residues compared with the reaction ^{244}Pu(^{48}Ca,xn)$^{292-x}114$. However, the latter was not observed in our experiment.

On the other hand, the difference in the cross sections of the reactions with ^{244}Pu nuclei can be a consequence of a strong increase of the dynamic hindrance with the increase of the projectile charge and mass when changing from ^{48}Ca to ^{58}Fe projectiles. We cannot exclude that this factor is of major importance, although nonobservation of element 120 makes it difficult to evaluate quantitatively the hindrance on the formation of the nuclei of element 120 in the reaction ^{244}Pu + ^{58}Fe.

Further attempts to synthesize element 120 in this reaction would require an increased sensitivity of the experiment. To enhance the production of element 120, the choice of a more mass-asymmetric reaction like ^{248}Cm + ^{54}Cr (or even ^{249}Cf + ^{50}Ti) would be preferable.

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