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Investigation of the reaction $^{64}Ni + {}^{238}U$ being an option of synthesizing element 120

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ABSTRACT

This study is concerned with the search for entrance channels suitable to synthesize elements with $Z > 118$. Mass–energy distributions as well as capture cross-sections of fission-like fragments have been measured for the reactions $^{64}Ni + {}^{238}U \rightarrow {}^{302}120$ and $^{48}Ca + {}^{238}U \rightarrow {}^{286}112$ at energies near the Coulomb barrier. Compound nucleus fission cross-sections were estimated from the analysis of mass and total kinetic energy distributions. The cross-section drops three orders of magnitude for the formation of the compound nucleus with $Z = 120$ obtained in the reaction $^{64}Ni + {}^{238}U$ compared to the formation of the compound nucleus with $Z = 112$ obtained in the reaction $^{48}Ca + {}^{238}U$ at an excitation energy of the compound nucleus of about 45 MeV. From our analysis it turns out that the reaction $^{64}Ni + {}^{238}U$ is not suitable for the synthesis of element $Z = 120$.

1. Introduction

The existence of the island of stability in the region of nuclei with $Z = 114$ and $N = 184$ predicted theoretically [1] has induced an extensive experimental investigation in the field of superheavy element (SHE) synthesis. A considerable success was achieved in reactions of actinides with a double magic ^{48}Ca beam at FLNR [2] where the synthesis of SHEs with atomic number Z up to 118 has been claimed. Experimental data confirm the theoretical prediction of the increase of the half-lives following the increase of the neutron number of the compound nucleus [2]. Unfortunately, the isotopes of SHE formed in these ^{48}Ca induced reactions cannot reach the neutron closed shell with $N = 184$ due to the lack of 7–9 neutrons.

Nuclei with $Z > 118$ cannot be synthesized in ^{48}Ca induced reactions since ^{249}Cf is the heaviest target material available for these purposes. A possible alternative pathway is represented by the complete fusion of ^{238}U, ^{244}Pu and ^{248}Cm nuclei with heavier projectiles such as ^{58}Fe or ^{64}Ni leading to the formation of compound nuclei (CN) with $Z = 118$–124 and $N = 178$–188.

Since at energies near the Coulomb barrier the fusion reactions between two heavy nuclei are strongly hindered by competing quasi-fission (QF) [3–6] and deep-inelastic reactions, more detailed experimental studies of the reaction mechanism are required to provide realistic estimates of the probability of producing compound nuclei in such reactions, especially in connection with the entrance channel properties. The most neutron rich isotope of element $Z = 120$ with $N = 182$ may be synthesized in three different

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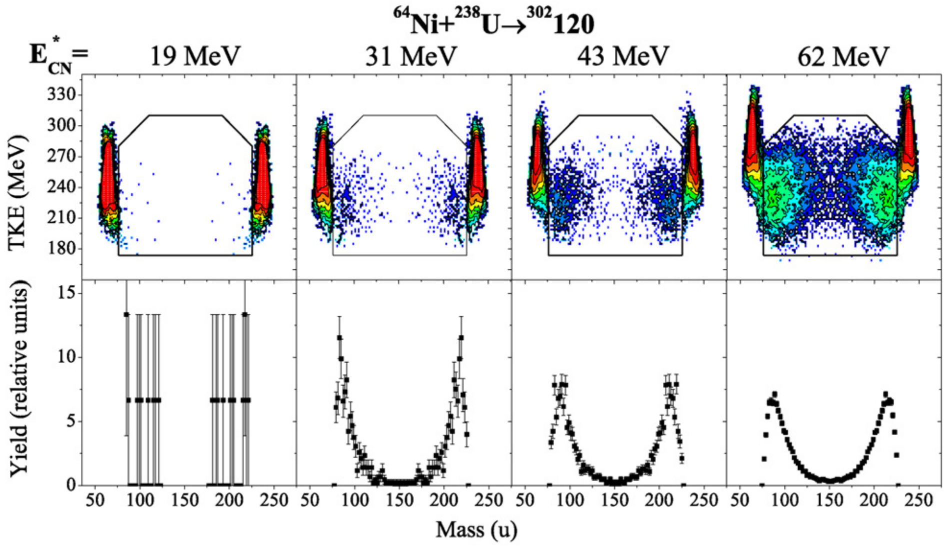

Fig. 1. (Color online.) Two-dimensional TKE-mass matrices (upper panels) and yields of fragments inside the contour lines in the TKE-mass matrices (bottom panels) measured in the $^{64}Ni + ^{238}U$ reaction at projectile energies of 330, 343, 358 and 382 MeV corresponding to excitation energies of the CN of 19, 31, 43 and 62 MeV, respectively.

fusion reactions: $^{54}Cr + ^{248}Cm$, $^{58}Fe + ^{244}Pu$ or $^{64}Ni + ^{238}U$. The reaction $^{54}Cr + ^{248}Cm$ is more favorable due to its larger mass asymmetry in the entrance channel [7]. However, some gain in the fusion cross-section for the $^{64}Ni + ^{238}U$ reaction may be caused by the lower excitation energy at the Bass barrier [8] compared to the other two reactions.

The main aim of the present study is to evaluate the reaction $^{64}Ni + ^{238}U$ as a possible candidate for the synthesis of the element with $Z = 120$. This Letter is concerned with the results of experimental studies of the properties of binary products in reactions of the magic projectiles ^{48}Ca and ^{64}Ni with the same target ^{238}U at energies around the Coulomb barrier. The mass–energy distributions of binary fragments as well as their cross-sections have been measured. The reaction $^{48}Ca + ^{238}U$ has been performed in order to test a data analysis methodology to further apply it to the data of the reaction $^{64}Ni + ^{238}U$. The method is an attempt to extract estimates of the fusion cross-section from the TKE distribution for fixed fragment mass intervals.

2. Experiment

Two separate experiments were performed: $^{64}Ni + ^{238}U$ at the Physics Department of the University of Jyväskylä using a ^{64}Ni beam from the cyclotron K-130, and $^{48}Ca + ^{238}U$ at the Laboratori Nazionali di Legnaro, using a ^{48}Ca beam from the XTU Tandem-ALPI accelerator complex. The beam energy ranges were $E_{lab} = 320$–385 MeV in the case of ^{64}Ni with a resolution of about 1%, and 220–260 MeV in the case of ^{48}Ca with a resolution of about 0.2%. Beam intensities on the targets were 1–2 pnA. The targets were built by evaporation of metallic ^{238}U (400 μg/cm^2) and $^{238}UF_4$ (100 μg/cm^2) on carbon backings (28–50 μg/cm^2). In both cases the enrichment was 99.99%. During the experiment the carbon backing faced the beam.

Binary reaction products were detected by the two-arm time-of-flight spectrometer CORSET [9]. Each arm of the spectrometer consisted of a compact start detector and a position-sensitive 9×7 cm^2 stop detector, both based on microchannel plates. The arms of the spectrometer were positioned at angles $+64°$ and $-64°$ to the beam axis for the reactions with ^{48}Ca and $+60°$ and $-60°$ for the reactions with ^{64}Ni. With this choice of angles, the scission axis is orthogonal to the beam axis for the case of symmetric splitting in both reactions. In other words, the fragments are detected at 90° in the center of mass frame. The distance between start and stop detectors was 15 cm. Start detectors were placed at a distance of 5 cm from the target. The angular acceptance for both arms was $\pm 12.5°$ in-plane and $\pm 10°$ out-of-plane. A typical mass resolution of the spectrometer in these conditions is about 2–3 u.

Four silicon detectors, placed above and below the reaction plane, and to the left and right of the beam at the same scattering angle of 16° were used to monitor the beam intensity and position continuously and also to normalize the yields to cross-sections.

The data processing assumes standard two-body kinematics [9]. Fragment energy losses in the target, the backing and the start detector foils were taken into account. Special attention was paid to the folding angle correlations both in and out of the reaction plane, and only events corresponding to a two-body process with full linear momentum transfer were considered.

3. Results and discussion

Figs. 1 and 2 display the measured TKE-mass distributions of binary fragments of the reactions $^{64}Ni + ^{238}U$ and $^{48}Ca + ^{238}U$, respectively. In the TKE-mass matrix the reaction products with masses close to those of the projectile and target are identified as quasi-elastic and deep-inelastic events, and were not considered in the present analysis. Reaction products lying between quasi-elastic peaks are assumed as totally relaxed events, i.e., as fission (or fission-like) fragments. We have surrounded those events by solid lines in the TKE-mass distributions, and their respective mass distributions are presented in the bottom of Figs. 1 and 2.

Mass–energy distributions of both of the studied reactions have the typical wide two-humped shape caused by QF under the influence of closed shells with $Z = 82$ and $N = 50$, 126. In the case of the $^{48}Ca + ^{238}U$ reaction the maximum yield corresponds to fragments with heavy masses 208 u, while for the reaction $^{64}Ni + ^{238}U$ the maximum yield corresponds to fragments with heavy masses 215 u. Based on the simple assumption of an N/Z equilibration, the nuclear shells with $Z = 82$ and $N = 126$ correspond to heavy fragment masses 207–209 u for both of the reactions. The neutron shell at $N = 50$ results in a light fragment of mass 82–83 u in the reaction with ^{48}Ca-ions as well as with ^{64}Ni-ions, but the complementary heavy masses for this nuclear shell are different: 204 u and 219 u, respectively. Thus, the major part of the asymmetric

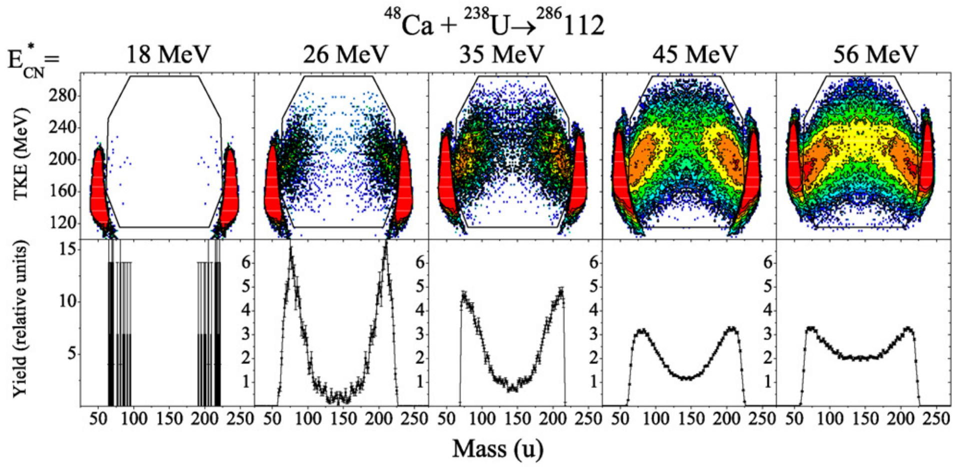

Fig. 2. (Color online.) Same as Fig. 1, but for the $^{48}Ca + ^{238}U$ reaction at projectile energies of 212, 222, 232, 244 and 258 MeV corresponding to excitation energies of the CN of 18, 26, 35, 45 and 56 MeV, respectively.

QF peak fits into the region of the $Z = 82$ and $N = 126$ (double magic lead) and $N = 50$ shells, and the maximum of yield of the asymmetric QF component is a mixing between all these shells. In the formation of the asymmetric QF component the closed shell at $N = 50$ seems to be effective together with the shells $Z = 82$ and $N = 126$ and leads to the shift of the asymmetric QF peak from mass 208 u to 215 u at the transition from ^{48}Ca ions to ^{64}Ni ions.

The dispersions of asymmetric QF fragment mass distributions increase as the projectile energies increase. At the lowest excitation energy of the CN of ~ 18 MeV one can see only asymmetric QF fragments for both reactions. The yield of symmetric fragments increases with increasing excitation energy as well, but the growth is less in the reaction $^{64}Ni + ^{238}U$.

A guideline for the interpretation of the pattern following from the TKE-mass distributions comes from dynamical models. A realistic description of the mass, energy and angular distributions of the reaction fragments formed in deep inelastic scattering, QF and compound nucleus fission (CNF) processes in low-energy heavy ion collisions was performed in [10] by using Langevin type dynamic equations of motion. It was shown that the multi-dimensional adiabatic potential energy surface (calculated within the two-center shell model) plays the most important role in such processes. Fig. 3 shows the potential energy surface as a function of the mass-asymmetry and elongation for the nuclear system consisting of 120 protons and 182 neutrons (for more details on these calculations see Ref. [7]). This potential energy surface is strongly modulated by shell effects and leads to the appearance of deep valleys corresponding to the formation of well bound magic nuclei. In accordance with these calculations, at least three paths leading to the formation of fission-like fragments can be distinguished: (1) asymmetric QF (QF_{asym} in Fig. 3) caused by the influence of proton shells with $Z = 28$, 82 and neutron shells with $N = 50$ and 126; (2) symmetric QF (QF_{sym} in Fig. 3) determined by the shells with $Z = 50$ and $N = 82$; (3) CNF (CNF path in Fig. 3) leading to the formation of symmetric fragments. Thus, in the reaction $^{64}Ni + ^{238}U$ the mass-symmetric fragments may be formed by different modes: either as a result of CNF or symmetric QF processes or as a tail of asymmetric QF process. It can be shown that the same pattern holds for the $^{48}Ca + ^{238}U$ reaction.

From Fig. 3 one can see that indeed the contact configuration of the less asymmetric combination $^{64}Ni + ^{238}U$ is located lower in the potential valley with respect to the partners $^{54}Cr + ^{248}Cm$ and $^{58}Fe + ^{244}Pu$, in particular in the proximity, but slightly after, the

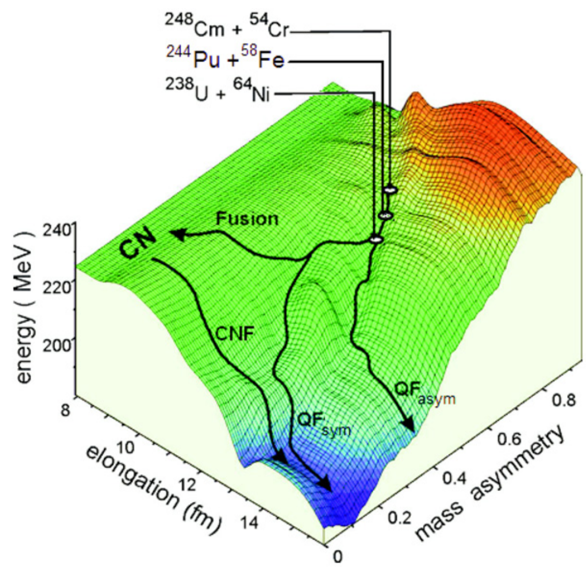

Fig. 3. (Color online.) Potential energy surface for the nuclear system consisting of 120 protons and 182 neutrons. Injection configurations (contact points) for the $^{54}Cr + ^{248}Cm$, $^{58}Fe + ^{244}Pu$ and $^{64}Ni + ^{238}U$ reactions are shown by the circles. Thick curves with arrows show schematically QF and fusion (CN formation) trajectories.

bifurcation point between the QF_{asym} and QF_{sym} paths. This means that in the case of $^{64}Ni + ^{238}U$ the nuclear system might be driven more toward the QF_{asym} channel than in the other two reactions. Consequently, a relatively higher contribution from the QF_{asym} path can be reasonably expected.

In Fig. 4a the TKE distributions of the fragments in the mass region $A_{CN}/2 \pm 20$ u are presented for the reaction $^{48}Ca + ^{238}U$ at $E_{lab} = 232$ MeV and for the reaction $^{64}Ni + ^{238}U$ at $E_{lab} = 358$ MeV. It is readily seen that both TKE distributions have a complex structure which is not consistent with only CNF. In fact, it is known that in such a case the average TKE of the partner fragments is substantially independent on the excitation energy and shows a typical Gaussian-like shape. By using the theoretical work of Ref. [10] as a guideline, we decompose each TKE distribution as a sum of three Gaussians. We use the systematics [11,12] as a starting point to evaluate mean and variance of the CNF mode. After a 3-Gaussian fitting procedure we can evaluate the cross-sections due to each of the three components. To test this approach, we apply this method

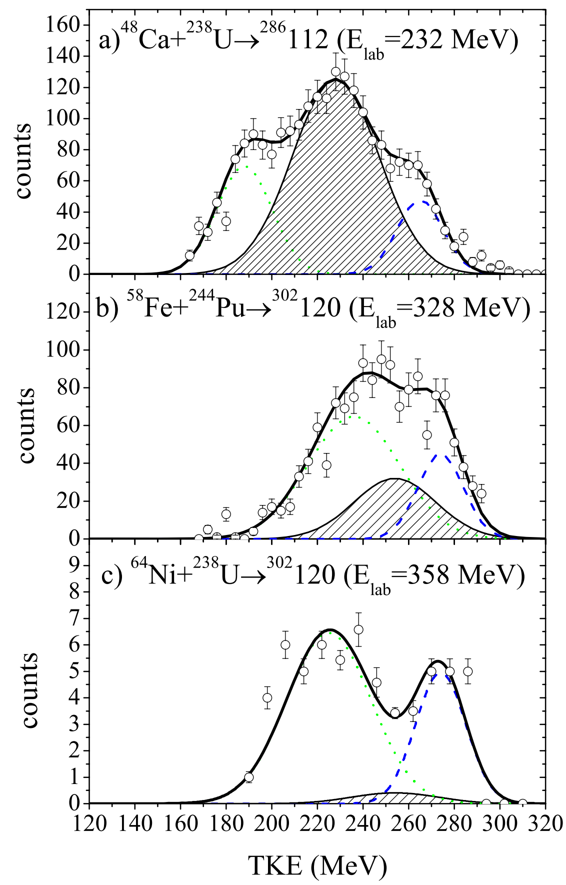

Fig. 4. (Color online.) TKE distribution of fragments with masses $A_{CN}/2 \pm 20$ u for the reaction $^{48}Ca + ^{238}U$ (a), $^{58}Fe + ^{244}Pu$ (b) and $^{64}Ni + ^{238}U$ (c). The open circles are the experimental points, the hatched region corresponds to CNF with energies taken from the Viola systematic, dashed and dotted curves represent high and low energy components of the TKE distribution.

to the reaction $^{48}Ca + ^{238}U$ where the capture, QF and CNF cross-sections have been measured by other authors [4,5]. Once this method is applied to the TKE distribution of fragments from the reaction $^{48}Ca + ^{238}U$, the degree of agreement of the estimated cross-sections with known experimental data provide us with the necessary confidence to apply the same method to the reaction $^{64}Ni + ^{238}U$.

From the Viola systematics we infer that the average TKE is in a first approximation a linear function of the Coulomb parameter $Z^2_{CN}/A^{1/3}_{CN}$ whereas from the systematics in Ref. [11] we can estimate the variance of the TKE distribution. For the $^{286}112$ CN ($Z^2/A = 43$) the variance σ^2 of the TKE distribution of CNF is about 400 MeV^2 [11] and the TKE is 233.7 MeV [12]. From the 3-Gaussian fit we obtain mean TKE values and standard deviation σ as shown in Table 1.

The TKE value of 228 MeV (Table 1) for CNF mode turns out to better agree with the older Viola systematics [13] which predicts a mean TKE of 226 MeV. However, we must consider that in the newer systematics [12] the data from Ref. [4] measured at energies well above the Coulomb barrier were included. In these reactions a considerable amount of QF processes contributes to the fission products. It turns out that the old systematics of Viola [13], without taking into account the experimental data from [4], provides a lower value of TKE better in agreement with our result. The standard deviation σ_{TKE} of the CNF component is around 20 MeV and also agrees well with the predicted value for the CNF process from the systematics [11].

Table 1
TKE decomposition for the $^{48}Ca + ^{238}U$ reaction at $E_{lab} = 232$ MeV.

	Mode	TKE (MeV)	σ_{TKE} (MeV)	Yield (%)
1-Gaussian	QF_{asym}	188.0 ± 3.0	14.6 ± 2.3	21 ± 5
2-Gaussian	CNF	228.5 ± 1.5	20.5 ± 2.5	68 ± 7
3-Gaussian	QF_{sym}	265.9 ± 1.9	8.6 ± 2.0	10 ± 4

The systematic errors of the TKE measurements are about ±2 MeV for the region of symmetric fragments.

Table 2
TKE decomposition for the $^{64}Ni + ^{238}U$ reaction at $E_{lab} = 358$ MeV.

	Mode	TKE (MeV)	σ_{TKE} (MeV)	Yield (%)
1-Gaussian	QF_{asym}	225.0 ± 3.0	25.0 ± 4.3	74 ± 13
2-Gaussian	CNF	252.1^*	23.0^*	$\leqslant 5$
3-Gaussian	QF_{sym}	278.0 ± 3.0	8.9 ± 2.5	21 ± 7

* Values were fixed according to [13] and [11].

Given the considerable good agreement with the systematics, we associate the 2-Gaussian component to the CNF process. Since the asymmetric fragments have lower TKE than the symmetric ones, the low energy component of the experimental TKE distribution may be associated with fragments originating from the asymmetric QF process. The high energy part may arise instead from the symmetric mode of the QF process. Furthermore, we note that the mean TKE from the QF_{sym} mode is about 40 MeV higher than the mean TKE for the CNF mode. Considering that both QF_{sym} and CNF modes give rise to symmetric mass fragments, the difference in mean TKE can be taken as an evidence that in the QF process a complete dissipation of the entrance channel energy does not occur. As a consequence, the symmetric fragments with high TKE do not originate from complete fusion because the final fragments retain part of the entrance channel total kinetic energy.

In contrast to the $^{48}Ca + ^{238}U$ reaction, for the reaction $^{64}Ni + ^{238}U$ the TKE distribution has more pronounced low and high energy components (see Fig. 4c), while the component with an average TKE value of 252 MeV, corresponding to the old Viola systematics, is highly reduced. A 3-Gaussian fit, performed according to the method used for $^{48}Ca + ^{238}U$, provided the means and variances shown in Table 2. Because of the lower statistics, we fixed the mean and variance of the CNF component to the values predicted from the systematics [13] and [11], respectively. Only an upper value for the relative yield of the CNF component can be reasonably given.

To evaluate the integrated cross-section for each component we need to make an assumption about the angular distribution in the center of mass frame. The absolute differential cross-sections for all fission-like events observed in the reactions were measured at an angle $\theta_{c.m.} = 90°$ and at energies from well below to well above the Coulomb barrier. The capture cross-section σ_{cap} for the production of all fission-like events (sum of CNF and QF processes) were estimated assuming that the angular distribution is proportional to $1/\sin\theta_{c.m.}$. This procedure seems to be the most reasonable and is applied since detailed angular distributions are not available at present. In the estimate of the QF cross-section we took into account a correction due to the overlapping of fission-like events with those corresponding to deep-inelastic and quasi-elastic processes and cut off a part of the asymmetric fission-like fragments in the case of the Ni-induced reactions.

The obtained capture cross-sections as well as the cross-section for the formation of symmetric fragments with masses $A_{CN}/2 \pm 20$ u are presented in Fig. 5 for both reactions. The capture cross-sections for the $^{48}Ca + ^{238}U$ and $^{64}Ni + ^{238}U$ agree well with previous measurements [4,5]. Moreover, for the reaction $^{64}Ni + ^{238}U$ at

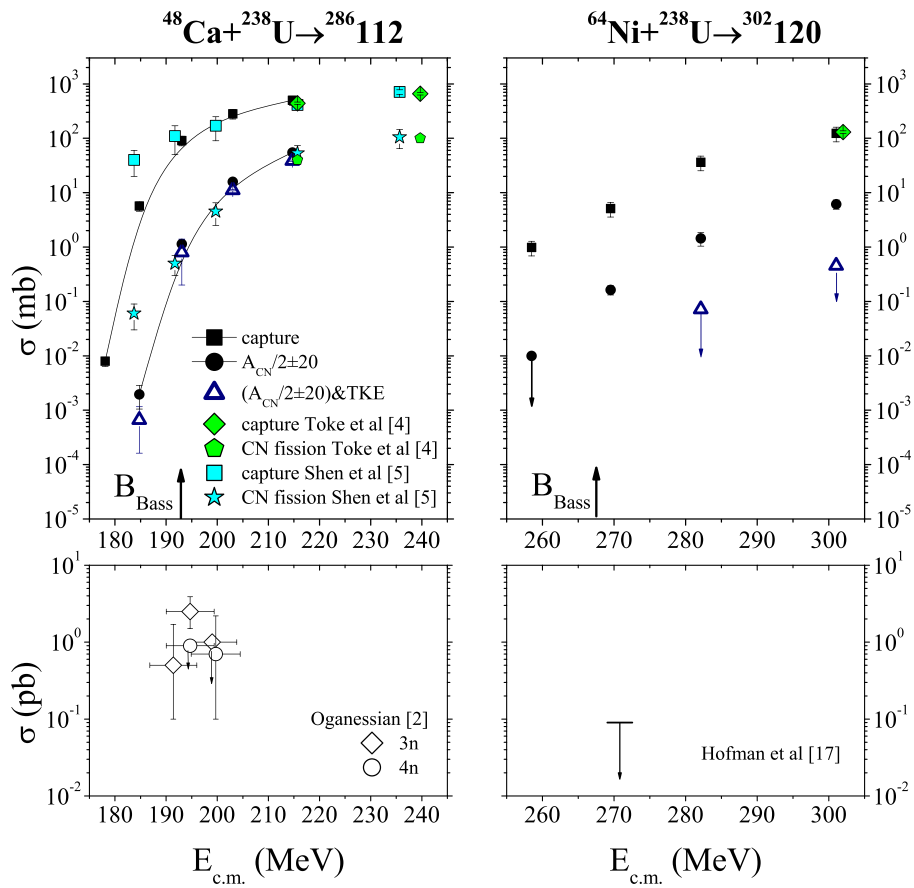

Fig. 5. (Color online.) Capture cross-sections (solid squares), cross-sections for formation of fragments with masses $A_{CN}/2 \pm 20$ u (circles) and with the restriction of TKE corresponding to the Viola systematic (open triangles) for the reactions ^{48}Ca, ^{64}Ni + ^{238}U. Open squares and rhombs represent the capture cross-sections, stars and pentagons represent the CNF cross-sections from [4,5]. Open rhombs and circles on the bottom panel are the evaporation residue cross-section from [2,17].

Table 3
Relative contributions of all symmetric fragments ($\sigma_{(A_{CN}/2\pm20)}$) and symmetric fragments with TKE corresponding to the older Viola systematics (σ_{CNF}) to the capture cross-section (σ_{cap}) for the ^{48}Ca + ^{238}U, ^{64}Ni + ^{238}U and ^{58}Fe + ^{244}Pu reactions at CN excitation energies of ∼ 45 MeV.

Reaction	$\frac{\sigma_{(A_{CN}/2\pm20)}}{\sigma_{cap}}$ (%)	$\frac{\sigma_{CNF}}{\sigma_{(A_{CN}/2\pm20)}}$ (%)	$\frac{\sigma_{CNF}}{\sigma_{cap}}$ (%)
^{48}Ca + ^{238}U	12 ± 2	68 ± 3	8 ± 4
^{64}Ni + ^{238}U	4 ± 1	$\leqslant 5$	$\leqslant 0.2$
^{58}Fe + ^{244}Pu	8 ± 3	$\leqslant 25$	$\leqslant 2$

$E_{lab} = 390$ MeV a total reaction cross-section of $\simeq 850$ mb was derived in [14] which is consistent with the value quoted in Ref. [4]. Subtracting from it the total transfer cross-sections, also measured in [14], amounting to $\sigma_{tr} \simeq 670$ mb, we obtain a "residual" or capture cross-section of $\sigma_{res} \simeq 180$ mb. This value compares well with 150 mb quoted in Ref. [4] and this work ($\sigma_{cap} = 122 \pm 40$ mb at $E_{lab} = 382$ MeV) and which is denoted as capture reaction (sum of CNF and QF).

Additionally, Table 3 gives the relative contributions of the symmetric fragments (with mass $A_{CN}/2 \pm 20$ u) to all fission-like events for the reactions ^{48}Ca + ^{238}U and ^{64}Ni + ^{238}U at an excitation energy of ∼ 45 MeV of the CN. The relative contribution of the symmetric fragments with TKE corresponding to the older Viola systematics is also presented in Table 3. Earlier the mass–energy distributions of fission-like products for the system ^{58}Fe + ^{244}Pu at $E_{lab} = 328$ MeV which lead to the same superheavy nucleus $Z = 120$ were measured in Ref. [15]. We have performed a similar analysis also for this system. The TKE distribution, measured under the same conditions, is shown in Fig. 4b and the relative yields are shown in Table 3.

In Fig. 5 the CNF cross-sections estimated in our ^{48}Ca + ^{238}U experiment are compared with the ones measured by other authors. This comparison is important to validate the ability of the procedure proposed to extract cross-sections from a 3-Gaussian fit to the TKE distributions. A good agreement with data in the literature would give us more confidence on the results when the same method is applied to the reaction ^{64}Ni + ^{238}U.

In the works [4,5], the angular distributions of fission-like fragments from the system ^{48}Ca + ^{238}U were measured and these measurements were taken into account to select the CNF (filled stars and pentagons in the left panel of Fig. 5). These data are in very good agreement with the data obtained in this work except for the energy points below the Bass barrier. The reason for this difference might be ascribed to the use of the reverse kinematics method [5] that leads to a worse mass resolution due to the larger velocity of the center of mass. As a result, the separation of fission-like events from elastic, quasi-elastic and deep inelastic scattering is more difficult to achieve.

On the basis of the reasonably good success of the analysis method proposed, we can draw some main conclusion. The capture cross-sections are about a few hundred millibarns for Ca and Ni induced reactions, whereas the formation of symmetric fragments is one order of magnitude less for the reaction ^{64}Ni + ^{238}U. Yet, in the case of the Ca + U at the highest energy, approximately 70% of the events have the TKE expected for the CNF process, whereas in the case of the ^{64}Ni + ^{238}U only a few percent of symmetric fragments have the TKE compatible with the Viola prediction for the ^{302}120

CNF. While the ^{64}Ni + ^{238}U reaction has lower excitation energy at center of mass energies close to the Bass barrier, the CNF cross-section is suppressed by stronger symmetric and asymmetric QF processes and the expected gain in CN survival probability was not observed.

The CNF cross-section in the $^{64}\text{Ni} + {}^{238}\text{U} \rightarrow {}^{302}120$ case drops three orders of magnitude with respect to the $^{48}\text{Ca} + {}^{238}\text{U} \rightarrow {}^{286}112$ case. This is unfortunately a limiting factor. Furthermore, the relative contribution of the CNF from ^{64}Ni + ^{238}U is much lower than in the case of $^{58}\text{Fe} + {}^{244}\text{Pu} \rightarrow {}^{302}120$. Recently the experiments aimed at the synthesis of isotopes of element $Z = 120$ have been performed using the $^{244}\text{Pu}(^{58}\text{Fe}, xn)^{302-x}120$ reaction [16] and $^{238}\text{U}(^{64}\text{Ni}, xn)^{302-x}120$ reaction [17]. A cross-section limit of 0.4 pb at $E^* = 44.7$ MeV for the former reaction and 0.09 pb at $E^* = 36.4$ MeV for the latter reaction were obtained. In the case of ^{48}Ca + ^{238}U reaction the evaporation residue cross-section for $3n, 4n$ channels is about a few pb. Thereby in the transition from Ca to Fe and Ni ions, the evaporation residue cross-section drops down at least one and two orders of magnitude, respectively.

Thus, we conclude that the reaction ^{64}Ni + ^{238}U is less favorable compared to ^{58}Fe + ^{244}Pu for production of the superheavy element with atomic number 120.

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