On the Possibility to Synthesize Superheavy Nuclei of Higher Neutron Numbers

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Abstract: The laboratory synthesis of nuclei of atomic numbers above Z = 110 having the maximum achievable neutron number N is considered. Cold fusion reactions are limited with monotonous decrease of the fusion cross section below 0.01 pb for Z = 114. The cross section of hot fusion reactions resulting in the synthesis of nuclei of elements of Z = 114, 115, 116, and 118 show no evidence of the influence of the predicted neutron subshell at N = 172. The limitations to create more neutron reach superheavy nuclei is analyzed in the case of radioactive beams with half-lives longer than 2 s. Theoretical and experimental half-life values for nuclei of Z = 110-118 are compared and half-life limitations are considered. New attempts to synthesize element of Z = 120 and 122 is discussed.

1. Introduction

In the middle of the 1960s nuclei of 12 elements beyond uranium were experimentally synthesized up to lawrencium (Z = 103) and rutherfordium (Z = 104) due to the stabilizing effect of the nuclear shell structure. At that time the question of the shell structure and its effect on nuclear stability was actually topical and the possibility of the existence of superheavy nuclei (SHN) was also considered [1]. It was proposed that in the case of closed proton and neutron shells the nuclei should have half lives long enough to be experimentally observed. Further theoretical studies led to the most probable closed proton shell at Z = 114 and closed neutron shell at N = 184 [2–4]. Later some model calculations led to other values of the closed proton shell at N = 184 has a relatively stabile position in the theory. An important step forward was made in 1967 – 68 by V.M. Strutinsky [5] presenting quantitative calculation of the microscopic part of the shell correction to the binding energy of heavy nuclei.

The first attempts to synthesize superheavy nuclei around Z = 114 were too optimistic. The first calculated half lives were of the order of 10^{8-9} years and at that time there were no reliable estimations of fusion reaction cross sections. The available experimental technique gave several basic possibilities for the synthesis: a) fragmentation reactions of two symmetric nuclei with similar proton numbers [7] and "gentle fusion" of two rare earth nuclei [6]; b) complete fusion reactions of the heaviest stabile double magic nucleus ²⁰⁸Pb and the neighbouring ²⁰⁹Bi with available stable beams of Z = 26-34 [8]. This type of complete fusion reaction led to the synthesis of nuclei of elements of Z = 107-113 [9–15]; c) the third possibility was the combination of as heavy transuranium target nuclei as available with beams of light nuclei, giving the desired Z. Yu.Ts. Oganessian realized a se-

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ries of experiments of this type of fusion reactions choosing the double magic nucleus of 48 Ca as projectile and synthesized the nuclei of elements of Z = 114, 115, 116, and 118 [16 20].

In fact the progress in the synthesis of nuclei with higher and higher Z was influenced by several factors. First of all the availability of the required beams and their intensity delivered from ion sources. The lack of reliable theoretical ideas about the process of fusion of heavy nuclei led to extremely large uncertainties in cross section and half life estimations and from here to many unsuccessful experiments.

2. Cross-section limitation

The production cross section of complete fusion reactions of heavy ions is a result of several physical processes playing role in this process – first of all prompt fission, deep inelastic scattering, complete fusion and compound nucleus survival probability. The process of fusion is influenced also with other effects, like the shell correction energy, nuclear spin, shape of the interacting nuclei and others.

There are several theoretical approaches to calculate the cross section of complete fusion reactions, but in general the reliability of these calculations is problematic. In cold fusion reactions of the double magic target nucleus of ²⁰⁸Pb and the neighbouring ²⁰⁹Bi with neutron reach Ti, Fe, Ni, Zn and similar ions the experimental cross section is steadily decreasing from 5 10 ⁷ barn for Z = 102 to 5 10 ¹⁴ barn for Z = 113 [15], i.e. the decrease of Z by one unit results in 6-fold decrease of the cross section in average as is shown in Fig. 1. No measurable influence of the stabilizing shell correction energy, isotopic spin or other parameters on the reaction cross section was observed. No explanation was find for example for the significant jump of the 1n channel cross section from 3.3 ($\frac{62}{27}$) pb for ²⁰⁸Pb(⁶²Ni, 1n) [12] to 15 ($\frac{9}{6}$) pb for ²⁰⁸Pb(⁶⁴Ni, 1n) [21] differing only by 2 neutrons in the Ni ions.

The hot complete fusion reactions, based on actinoid targets from uranium to californium and on the double magic ⁴⁸Ca projectile ions has an unexpected feature. In spite of theoretical predictions the cross section of all realized reactions have very similar values differing less than one order of magnitude as it is illustrated in Fig. 2. and Fig. 3. For Z =112 118 and for the neutron number N = 170 177 no systematic trend in measured cross section values was observed. It means no observable influence of the predicted neutron subshell at N = 172 [23] or the influence of the closed neutron shall at Z = 184. To draw serious consequences for the theory, these cross section values need to be confirmed in independent experiments and at higher reliability.

3. Target – projectile limitation

To investigate the region of superheavy elements above Z = 112, the only today known approach is the method of hot fusion reaction of actinoid target nuclei and suitable accelerated ions. The successful chain of reactions based on U, Pu, Cm, Bk, and Cf targets and stable ⁴⁸Ca ions is exhausted. In these reactions the limits of the radioactive target and stable projectile nuclei combinations were reached in the direction to more neutron rich nuclei towards to N = 184. To reach higher neutron numbers up to the predicted closed

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neutron shell at N = 184 actinoid target nuclei have to be bombarded with ions heavier that ⁴⁸Ca. But even in this case only at Z = 124 one can reach N = 184 as it is shown in Fig. 4.

In the region of interest (Z = 114-126) higher neutron numbers are achievable only using radioactive beams (RBs). Considering available actinoid target nuclei and radioactive beams of ions having half lives longer than 2 seconds, the achievable front line is shown in Fig. 4. Even in these case the predicted closed neutron shell at N = 184 is achieved at Z = 119. Today we have no idea how to reach the proposed closed neutron shall (N = 184) at Z = 114.

The idea to use radioactive beams to synthesize neutron reach superheavy elements is not a new one. The basic problem is the available intensity of such beams. Suitable neutron reach radioactive ions can be created by fragmentation of high energy nuclei and by consequent in-flight separation and deacceleration of the separated high energy ion to coulomb barrier energy level. With respect to the expected picobarn (10 40 m²) cross section level, the necessary beam intensity is of the order of 1 pµA or 10¹² ¹³ ions/s. The present approachable intensity of single ion RBs is below 10⁹ ions/s. To reach the 10¹² ¹³ ions/s level will be a very difficult task. New powerful ion source of primary beams, high energy accelerator to accept such beams, and probably the parameters of the fragment separator and deaccelerating ring should be specially adjusted to fulfill the task.

4. Half life limitation

The first phase of a heavy compound nucleus creation in a complete fusion reaction is a complex function of many internal and external parameters of both interacting nuclei. After the formation of the compound nucleus its survival probability depends on fewer internal parameters of the formatted compound nucleus itself. This gives for the theory the possibility to calculate more reliable half-life values than in the case of the fusion probability.

Half life calculations made by Sobiczewski et al. [24] show a clear dependence of alpha decay half lives on both, the proton number Z and the neutron number N of a particular nucleus. With increasing atomic number Z the half lives of all isotopes of the given Z are monotonously decreasing, but there are two significant peaks at neutron numbers around the proposed neutron subshell of N = 162 and closed neutron shell of N = 184 (see Fig. 5).

To check the reliability of these calculations for superheavy elements is rather problematic. First of all the theory gives data only for even-even nuclei, but in the matrix of 121 nuclei of Z = 110-120 and N = 166-176 only 9 even-even nuclei have experimentally determined alpha-decay half lives. The comparison of the available experimental and calculated alpha decay half lives are given in Tab. 1. The calculated values are mostly underestimated and differ from the experimental ones from several times to two orders of magnitude.

The second problem is the uncertainty in the experimental data due to very low statistics, in some cases only one or two recorded events. Š. ŠÁRO

5. Present attempts

At present new attempts are made to go further in SHN synthesis. The synthesis of Z = 120 element is on the program in two laboratories. In JINR Dubna the reaction of 244 Pu + 58 Fe $^{299}120 + 3n$ is going on [22] and at GSI Darmstdat [25] the reaction of 238 U + 64 Ni $^{299}120 + 3n$ is on the way. Both reactions are leading to the same compound nucleus

 302 120* and the 3n evaporation channel will create the same evaporation residue of 299 120. The expected alpha decay chain of 299 120 after one unknown member (295 118) will follow the path of already synthesized alpha decay nuclei - 291 116 287 114 283 112 279 110 as it is illustrated in Fig. 6.

The cross section of both reactions leading to ²²⁹120 is uncertain. The hot fusion synthesis of all nuclei of elements of Z = 112, 114, 116, and 118 were based on the interaction of the neutron reach double magic ⁴⁸Ca ions with transuranium target nuclei. If the closed shells in ⁴⁸Ca played a substantial stabilizing role in the process of fusion then the cross section of both reactions, leading to ²⁹⁹120 can fall significantly below 1 pb. The calculated alpha decay half life of ²⁹⁹120 is about 1 µs [24] or higher [26]. The time of flight of the evaporation residues from the target to the analyzing detector array is several µs, therefore the detection efficiency may be critical.

6. Perspectives

The cross section of the cold fusion reaction at Z = 113 is only 0.05 pb [15]. The monotonous decrease of from Z = 102 to Z = 113 predicts the expected value of s for $Z = 114 \ 0.01$ pb. This is below the acceptable beam time of several months to synthesize one nucleus of element Z = 114. There are expectations to design ECR ion sources delivering heavy ion beams of the order of 10^{14} ions/s which should allow to reach the 0.01 pb level at reasonable beam time. But at such a heavy ion beam intensity several secondary problems will appear. First of all the energy deposition in Pb or Bi targets is limiting at present the acceptable beam intensity to about 10^{12} ions/s. The second problem will be the background. The method of identification of new nuclei based on the alpha – alpha correlation method requires as low background count rate in the sensitive part of the energy spectra as possible. To avoid difficulties of this type special effort should by paid to the construction and shielding of all parts of the equipment from the target chamber to the detector array beyond the separator.

Properly designed transuranium targets will be able to accept beams of ions of the order of 10^{14} but the problem of the background will be serious also here, especially in the case of gas filed separators.

7. Conclusion

The artificial synthesis of nuclei heavier than uranium was, all the time, a front-line experiment requiring the most advanced laboratory equipment and novel physical approaches. The method used to synthesize the first transuranium nuclei exhausted its possibilities at mendelevium Md (Z = 101). The next generation of experiments postponed the frontier to siborgium (Z = 106). A new approach was needed to continue, the

cold fusion concept, which shifted the frontier to element 113, where the cross section of the 1n evaporation channel fall to 0.05 pb. An unexpected success of hot fusion reactions, based of the double magic ⁴⁸Ca ions and trasuranium targets pushed the frontier to element 118. The present attempts to go further are not based on novel physical ideas but on the expectation that the 3n hot fusion evaporation channel will work as well as in the case of the double magic ⁴⁸Ca ions. To get closer to the predicted closed proton shell at Z = 114, 120, or 126 and closed neutron shell at N = 184 new physical ideas and more advanced and powerful experimental technique should be involved.

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Table 1. Measured (upper data) and calculated (Sobiczewski et al. [24]) half-lives of the synthesizedelements of Z = 110118.

120						?
						1 2 ms
118						0.9 ms
						0.05 ms
116					15 ms	18 ms
						8 ms
114				160 ms	800 ms	
				8 ms	200 ms	
112			0.50 ms	101 ms		
			7 ms	1 ms		
110	9.7 ms					
	0.2 ms					
Z/N	166	168	170	172	174	176



Fig. 1. Experimental cross section for cold fusion reactions for evaporation residues of Z = 102 113 and 1n evaporation channel.





Fig. 2. Experimental cross section for hot fusion reactions for evaporation residues of Z = 112 118 and 3n, 4n evaporation channels [16 20].

Fig. 3. Experimental cross section for hot fusion reactions for evaporation residues of N = 170 177 and Z = 112 118 [16 20].



Fig. 4. Chart of superheavy nuclei. The full line represents the maximum possible number of neutrons in the particular nuclei, created in 3n evaporation channel hot fusion reactions of actinoid target nuclei and stable projectiles. The dashed line represents the same, but for radioactive projectiles having half-lives >2 s.

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Fig. 5. Alpha decay half life calculation for isotopes of heavy elements from fermium (Z = 100) to element of Z = 124 (after Sobiczewski et al [24].



Fig. 6. The expected decay chains of evaporation residues ²⁹⁹120 and ³⁰⁵122.

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