HEAVY - ELEMENT RESEARCH AT FLNR (Dubna)

Yu.Ts.Oganessian

Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research,
141980, Dubna, Moscow region, Russia

Abstract

Results of research on the synthesis and investigation of properties of heavy nuclei in the top of the Periodic chart of elements carried out by the FLNR (Dubna) - LLNL (Livermore) collaboration are presented.

These results have brought to the observation of a new region of nuclei stability near closed deformed shells Z=108, N=162 predicted by the macro-microscopic theory

The heaviest isotopes Z=104, 106, 108 and 110 have been synthesized in hot fusion reactions of ^{238}U, ^{244}Pu *and* ^{248}Cm *nuclei with ions of* ^{22}Ne *and* ^{34}S. *Their radioactive properties point to a substantial increase of stability with respect to spontaneous fission in the whole known region of nuclei of transfermium elements with* $N \leq 158$.

The partial half-life for the $^{262}104$ *nucleus with respect to, spontaneous fission has been experimentally determined:* $T_{sf} = 1.2^{+1.0}_{-0.5}$ *s. All other isotopes with* $Z \geq 106$ *undergo mostly the α-decay. The experimental systematics of the α-decay energies* $Q_\alpha(N)$ *for all the known isotopes with Z=98-110 determines the status and strength of structural effects near closed deformed neutron shells N=152 and N=102. It is demonstrated that the calculated values of nuclei near shells N=162 are in a good agreement with the experiment.*

The research has been carried out at the FLNR(JINR) on the beams of the U-400 heavy ion accelerator using the Dubna gas filled separator of recoils.

The discovery of a new region of nuclei stability near closed deformed shells allows to make more accurate assessments regarding the properties of heavier nuclides up to Z=120 and N=190. For them a much stronger effect from spherical shells Z=114 and N=180-184 is predicted which opens a possibility of further progress into the region of superheavy elements.

For the synthesis of isotopes of the element 114 with maximum neutron excess the $^{244}Pu(^{48}Ca,3,4n)^{289,288}114$ *hot fusion reaction is considered.*

Theoretical predictions

It is known that modern macro-microscopic theory as applied to the collective motion of heavy nuclei explains in general a number of experimental results: fission barrier configuration, shape isomerism, spontaneous fission half-lives, mass and energy distributions of fission fragments and as well as many other facts which have found no explanation in the classical liquid drop models.

The effects of the deformed nuclear structure appear to be most pronounced in the probability of spontaneous fission a process which actually defines the limit for the Periodic system of elements.

As a result of the fission barrier emergence, determined by the nuclear structure, partial spontaneous fission half-lives of heavy nuclei turn to be by 12-15 orders of magnitude larger than the values predicted by the classical liquid drop model of nuclei. Additional slowing down of the spontaneous fission of nuclei with the odd number of protons and/or neutrons makes this effect still more important.

Due to a high spontaneous fission stability isotopes of the heaviest synthesized elements undergo an alpha-decay. This circumstance, as it will be shown further, allows not only the synthesis of new nuclides but also a rather precise determination of their mass in the ground state, of the probability and energies of the decay.

Clearly, the theory can make certain predictions, in particular about the mass and radioactive properties of still unknown nuclei.

Such predictions were made in a number of papers. We are presenting here some data from papers by Z.Patyk and co-workers[1] where the masses and fission barriers as well as partial half-lives T_α and $T_{s.f.}$ of even-even nuclei with Z=100-112 and N=140-166 have been calculated.

Fig.1 presents a contour map of spontaneous fission half-lives as a function of proton and neutron numbers. Significant changes in $T_{s.f.}$ of nuclei far from the N=152 shell are determined to a great extent by another shell with N=162. It should be noted that both the neutron shells are referred to deformed nuclei in the ground state. The maximum stabilization against spontaneous fission is expected for the nucleus $^{270}108$(Z=108, N=162) for which the predicted $T_{s.f.}$ may reach 10^6- 10^9 s.

However, the calculation of spontaneous fission half-life $T_{s.f.}$ in the dynamical way consists of a search for a one-dimensional fission trajectory in a multi-dimensional deformation space, which minimizes the action integral corresponding to the penetration of the fission barrier. The trajectory may not be the only one, and it leads to different fission modes, which differ considerably in the collective motion time and, hence, in fission probability[2]. Although the calculated static barrier heights are almost equal, the differences in half-life estimates can be attributed to varying assumptions regarding the dynamical path through the fission barrier and consequent inertial mass.

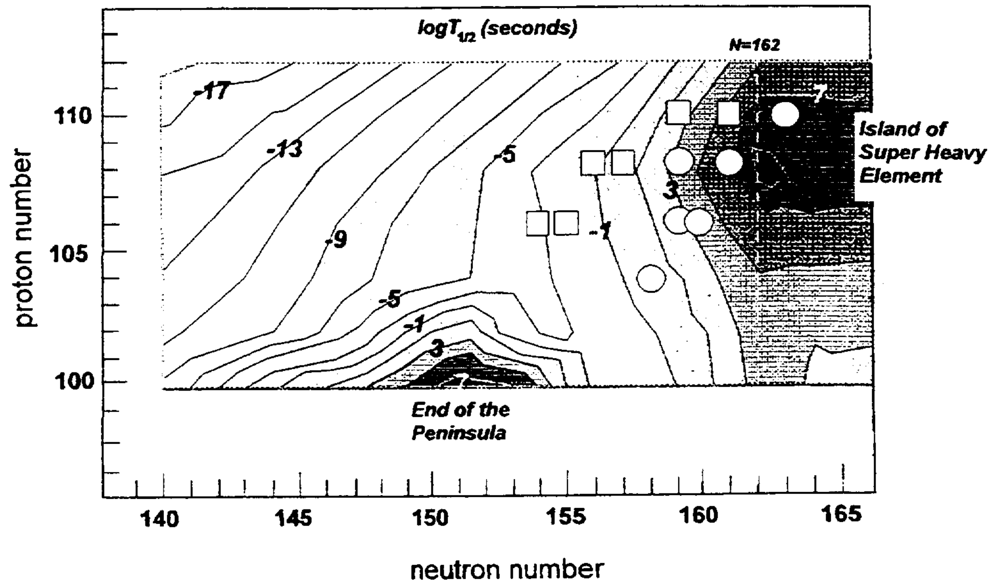


Fig.1 Contour map of the logarithm of spontaneous fission half-life T_{sf} (seconds) calculated by Z.Patyk et al. [1]. The difference in the values of $LogT_{sf}$ between neighbouring lines is 2. Squares: even-even isotopes produced in cold fusion reactions; circles: isotopes produced in hot fusion reactions.

For example, P.Möller et al.[3], taking ^{258}Fm as a model for heavier nuclei, assume that the path after the first barrier is short with the emerging fragments being nearly spherical and close to the doubly magic ^{132}Sn. On the other hand, Z.Patyk, et al[1]. calculate dynamical barriers that show a different path, higher inertial mass, and consequently much longer spontaneous fission half-lives. This competition between static and dynamical features of the spontaneous fission process leading to so large differences in stability makes experiments that explore ground-state decay properties of nuclei around N=162 and Z=108 one of the most important tasks in heavy element research.

Reactions of Synthesis

It is known that the heaviest elements of the Periodic table have been synthesized in the cold fusion reaction ^{208}Pb(HI,n). One of the main advantages of cold fusion is a relatively low excitation energy of the compound nucleus (E_x=10-20 MeV). At such a small excitation nuclear shell effects disappear, although not completely, which gives a certain stability to the system with respect to fission. The transition into the ground state occurs by emission of just one or two neutrons and γ-rays [4].

The production cross sections of the known most heavy nuclei are in the region of picobarns[5,6]. Despite of the obvious advantages of cold fusion reactions they lead to the production of isotopes of elements 106 and 108 with N=155 and 157 correspondingly, which are still far from the peak of the predicted island of stability.

In principle, a significant growth in the number of neutrons in evaporation residues (EVRs) can be obtained in fusion reactions between heavy actinide nuclei of the ^{244}Pu, ^{248}Cm type and projectiles such as ^{22}Ne, ^{26}Mg, ^{36}S. In such reactions the excitation energy of a compound nucleus even at the Coulomb barrier is more than 40 MeV. The structural effect practically disappears at such a high excitation energy; their fission barrier is determined only by the macroscopic (liquid drop) component of the nucleus deformation energy[7].

It is well known that for transactinide nuclei the fission barrier is practically equal to zero. In the absence of a fission barrier an excited nucleus becomes totally unstable to fission which should lead to a strong decrease in the probability of its transition to the ground state via a cascade evaporation of 4-5 neutrons. Under these conditions the survival of EVRs totally depends on the dynamic properties of the excited compound nucleus.

Investigation of excited nucleus fission dynamics by measuring the probability of pre-fission neutron emission [8,9] together with experimental data for the cross section of (HI,4-6n) channels obtained in the region Z_{CN}=102-105[11,12] makes it possible to compare the possibilities of sythesizing isotopes Z≥106 in cold and hot fusion reactions.

The expected channel cross section (HI,4-5n) values are slightly lower than in the cold fusion reactions. However, these difficulties are compensated by a possibility of obtaining isotopes of elements 106 and 108 with N=160 and 161 just near the deformed shells with Z=108 and N=162.

Observations of Enhanced Stability of Nuclei Z=104-110 Near Closed Shells

Essentially, this was the underlying idea of a joint JINR (Dubna) - LLNL (Livermore) experiment on the synthesis of element 106 heavy isotopes [14].

The ground state decay properties of ^{266}106 should be a quite sensitive probe of the theoretical predictions. If there is an increased stability near N=162 and Z=108, the isotope ^{266}106 should have a SF- or α-decay half-life of tens of seconds [15]. Otherwise, ^{266}106 should decay by SF with a half-life of ~100 μs [3]. A T_{sf} difference is of ~10^5 or more. Thus, a distinct signature for enhanced nuclear stability near N=162 and Z=108 would be the observation of the α-decay of ^{266}106 followed by the SF decay of the daughter nucleus ^{262}104. A signature for the odd-A isotope ^{265}106 would be the observation of its α-decay followed by sequential α-decays of the known nuclides ^{261}104 and ^{257}102.

To produce ^{265}106 and ^{266}106 we have used the complete fusion reaction ^{248}Cm+^{22}Ne at bombarding energies which are expected to provide maximum cross sections for the 4n and 5n evaporation channels.

At the irradiation of the ^{248}Cm target with a ^{22}Ne total ion beam doze of $1.6 \cdot 10^{19}$ produced on the U-400 accelerator (FLNR) by means of a gas-filled recoil separator two new most neutron-rich isotopes of element 106 with masses 265 and 266 have been synthesized.

Table I

Nuclide	Principal decay mode	Alpha-particle energy, MeV	Half-life
^{262}104	SF		$1.2^{+1.0}_{-0.5}$ s
^{265}106	α	8.63 to 8.91	2-30 s
^{266}106	α	8.63 ± 0.05	10-30 s
^{267}108	α	9.74 to 9.87	19^{+29}_{-10} ms
^{273}110	α	11.35	$0.3^{+1.4}_{-0.2}$ ms

Both the isotopes ^{265}106 (N=159) and ^{266}106 (N=160) undergo mostly the α-decay with energies E_α=8.71÷8.91 and 8.63 ± 0.05 MeV correspondingly. The energy of the α-decay of the even-even nucleus ^{266}106 (Q_α=8.76 MeV) determines its half-life T_α=10-30s. (see Table I).

Based on the six registered (α,sf) correlations referring to the α-decay of the ^{266}106 nucleus there was also determined the partial spontaneous fission half-life $T_{s.f.} = 1.2^{+1.0}_{-0.5}$ s of the daughter nucleus ^{262}104 (N=158).

Radioactive properties of even-even isotopes of ^{262}104 and ^{266}106 give an indication of a substantial growth of heavy nuclei stability to spontaneous fission when approaching the closed shells Z=108 and N=162 (Fig.2).

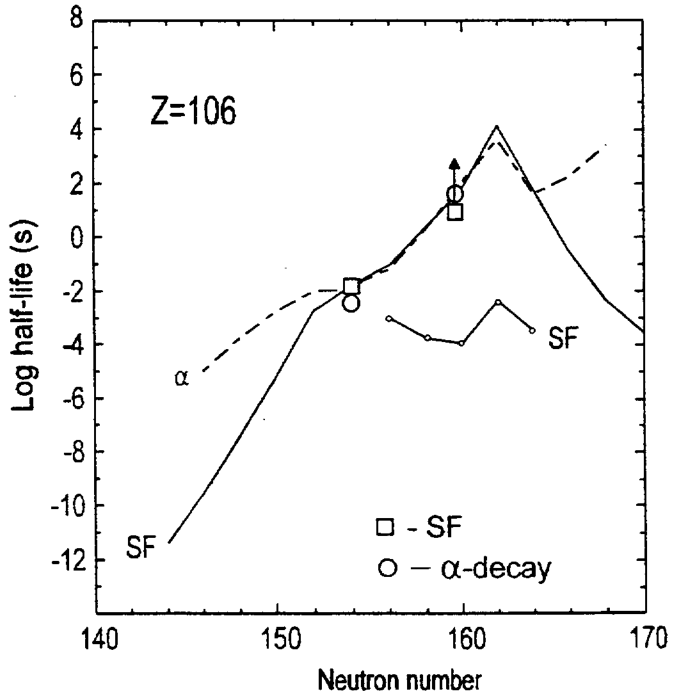


Fig.2 Partial half-lives predicted by A.Sobiczewski et al [15] for spontaneous fission and alpha-decay of even-even isotopes Z=106 shown by solid and dashed lines respectively. The solid line with points shows spontaneous fission half-life predictions by P.Moller et al. [3]. The experimental values for ^{260}106 [16,17] and results for ^{266}106 [14] are shown for comparison.

And really, with decreasing number of neutrons in the nucleus of $^{262}104$ by 6 units ($^{256}104$) or of protons - by 4 units (^{258}Fm), the spontaneous fission half-life gets correspondingly 250 and 2500 times shorter.

Similarly to this at the decrease of the number of neutrons in the nucleus of $^{266}106$ by 6 units ($^{260}106$) or of the number of protons by 4 units ($^{262}102$) the half-life of spontaneous fission becomes over 5000 times smaller. Moreover, a heavier nucleus has a larger half-life: $T_{s.f.}$=(Z=106, N=160) >>$T_{s.f.}$(Z=104, N=158).

This means that the nuclei obtained in this experiment are in the process of an abrupt increase of stability to spontaneous fission, as predicted by the macro-microscopic calculations by A.Sobiczewski et al.[15], (Fig.3). Another spontaneous fission mode, characterized by a short path of tunneling through the fission barrier [3] and leading to a sharp decrease of $T_{s.f.}$ for $^{266}106$ is suppresed by more than 10^4 times.

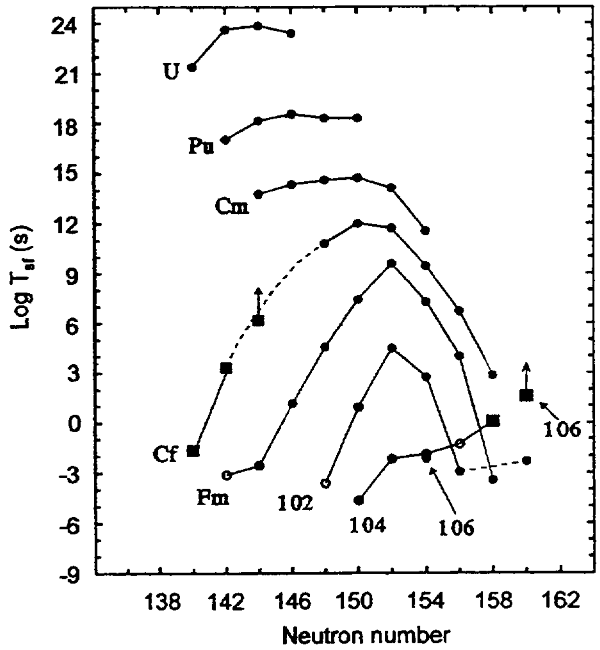


Fig.3 Spontaneous fission half-lives vs.neutron number for even-even isotopes transuranium elements. Squares: T_{sf} of isotopes of $^{262}104$ and $^{266}106$ produced in the $^{248}Cm(^{22}Ne,4n)$ reaction [14] and of light isotopes of Cf from the reactions $^{206\text{-}208}Pb(^{34}S,2\text{-}4n)$ [18].

Among all possible target-ion combinations leading to the production of a $^{270}108$ nucleus with closed shells Z=108 and N=160, the reaction $^{238}U(^{36}S,4n)^{270}108$ seems to be the most promising one. However, with account of the ^{36}S isotope high cost, experimenters in Dubna in March-April 1994 were using a beam of a more abundant isotope ^{34}S enriched to 90%.

At the irradiation of a ^{238}U target with a total ^{34}S-beam doze of $1.7 \cdot 10^{19}$, the position-sensitive strip detectors of recoils located in the focal plane of the separator registered 3(α-α) correletion events clearly pointing to the production of a new isotope of element 108 with a mass of 267[19,20].

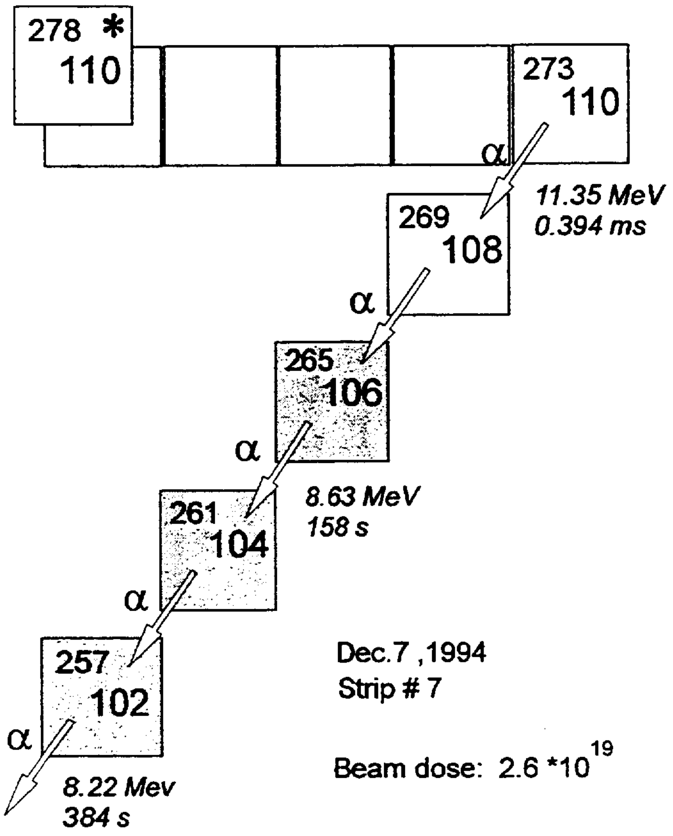
As seen in Table I, some 20-30 ms after the recoil nucleus entry into the detector its decay was observed with emission of an α-particle with an energy E_α=9.74-9.88 MeV followed by the subsequent decay of $^{259}104$ or $^{255}102$ daughter nuclei, the decay properties of which are well known.

The yield of $^{267}108$ nuclei in the 5n-channel (E_x=50 MeV) corresponds to the reaction cross section of about 3 pb (with a factor of 3 accuracy).

Finally, in September-December 1994, experiments on the synthesis of element 110 were performed. Three sequential α-decays pointing to of the formation of a new isotope of element 110 with A=273 were registered by the position-sensitive detectors situated in the focal plane of the separator during the exposure of a ^{244}Pu-target to a total beam doze of 2.7 10^{19}. The energy of the projectile ions was 190 MeV ($E_x \approx 50$ MeV) near the maximum of the 5n- evaporation channel [21,22].

As seen from Fig.4, approximately 0.4 ms after the moment the recoil nucleus had passed through the time of flight system and entered into the position sensitive strip detector there was observed its decay with emission of an alpha-particle with an energy E_α=11.35 MeV.

This decay followed by the subsequent decay of the grand-daughter nucleus $^{265}106 \xrightarrow{\alpha} {}^{261}104 \xrightarrow{\alpha} {}^{257}102 \xrightarrow{\alpha} {}^{253}$Fm previously known from the ^{248}Cm(^{22}Ne,5n)^{265}106 reaction: [14].



Dec.7 ,1994
Strip # 7

Beam dose: 2.6 *10^{19}

Fig.4 Decay chain of the nucleous ^{273}110 produced in the reaction ^{244}Pu+^{34}S.

The random coincidence of such correlation is $4 \cdot 10^{-4}$. The yield of the new nuclide in the 5n-channel (E_x=50MeV) corresponds to the reaction cross section of about 0.4 pb.

The experimental data of the reaction ^{244}Pu+^{34}S are still being processed, however the already obtained results are of certain interest.

The Q_α(N) dependence for all known even nuclei with Z≥98 is presented in Fig.5.

Irregularities in the behaviour of Q_α(N), as is well known, depend on the nuclear structure in the ground states. They are maximum near closed shells, where the Q_α value varies considerably. For example, for Po isotopes the difference in the value of Q_α between two even-odd nuclides: ^{209}Po(N=125) and ^{211}Po(N=127) -crossing shell N=126 is more than 2.5 MeV. Owing to the spherical shells the isotopes of lead and bismuth with Z=82 and N=126 are stable. For deformed shells with Z=102 and N=154 an analogous procedure gives a value ΔQ_α which is just about 0.1 MeV. However, the presence of shells effect in the deformed nuclei leads to a comparatively high stability of heavy actinide isotopes with a strong inhibition for spontaneous fission.

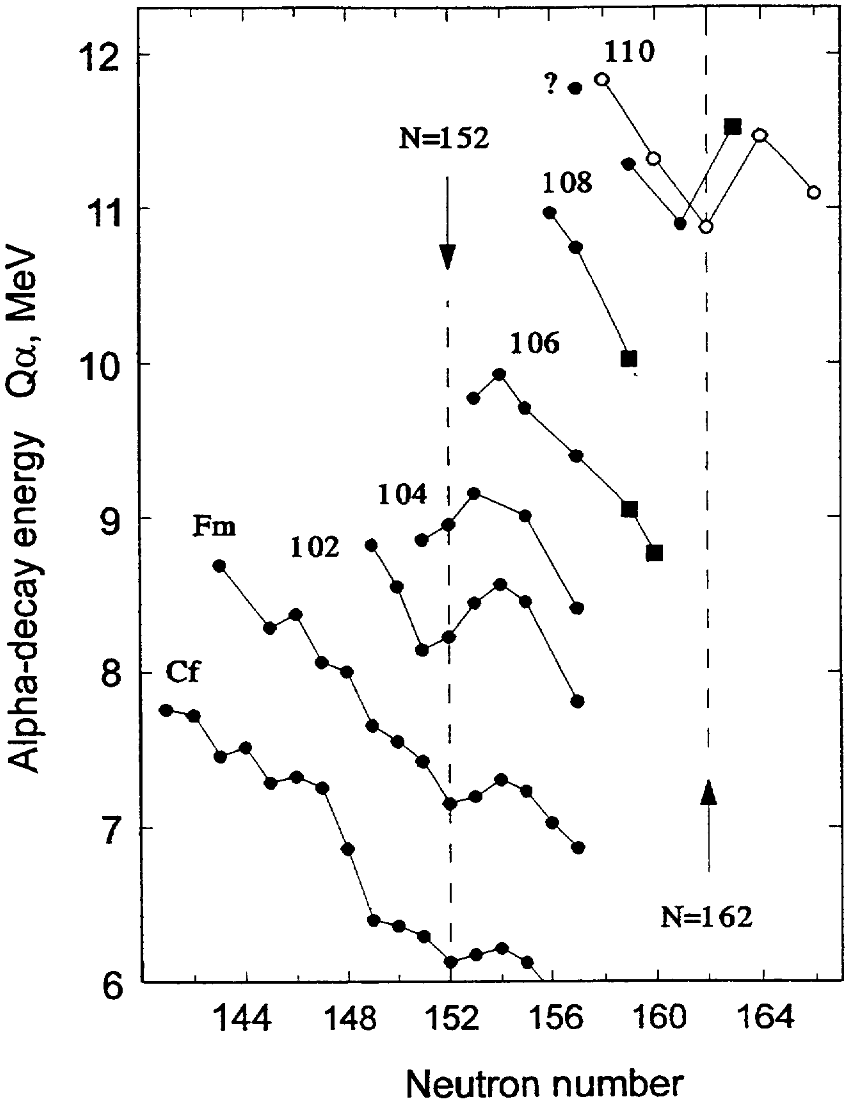


Fig.5 Alpha-decay energy vs. neutron number for isotopes of elements Z=98-110. Squares - experimental data for heaviest isotopes of the elements Z=104-110 produced in hot fusion reactions. Open circles - theoretical predictions for even-even isotopes Z=110. Deformed neutron shells N=152 and N=162 indicated by arrows.

Finally, from the results of the latest experiments on the element 110 synthesis performed at GSI for the isotopes of element 110 with N=159 and N=161 [23,24] and also from the observed decay event of the nucleus with N=163 it follows that ΔQ_α=0.65 MeV, which is close to what has been predicted by calculations [25].

Note that this comparison is carried out for two even-odd nuclei which are formed directly after the deexcitation of the compound nucleus. A more precise value of ΔQ_α for the ground states of these nuclei can be determined if they themselves are the products of the α-decay of the mother nuclei $^{275}112$ and $^{277}112$.

Experimental results on the synthesis of heavy isotopes with Z=104, 106, 108 and 110 confirmed the theoretical predictions about the substantial rise of nuclear stability in the region of deformed shells with Z=108 and N=162. The quantitative agreement with the experimental results has been achieved by means of slight variations of calculation parameters and, upon the whole, has been in agreement with the main postulates of the macro-microscopic theory about the great influence of the nuclear shells on the superheavy nuclei stability.

Towards Spherical Shells Z=114 and N≈180-184.

Formally, theoretical calculations of the masses and nuclear deformations may be continued for the determination of the radioactive properties of superheavy nuclides, in particular, of the nuclei near the expected spherical shell Z=114 and N=184.

The work by R.Smolanczuk and A.Sobiczewski[25] has shown that, provided there is a neutron excess in the heavy nuclei, the shell correction amplitude is still considerable (~6-7 MeV) for a wide range of nuclei, up to Z=120 and N=190. This is determined by the spherical shell Z=114, N=180-184, which embraces a wide range of superheavy nuclides of high-stability with respect to

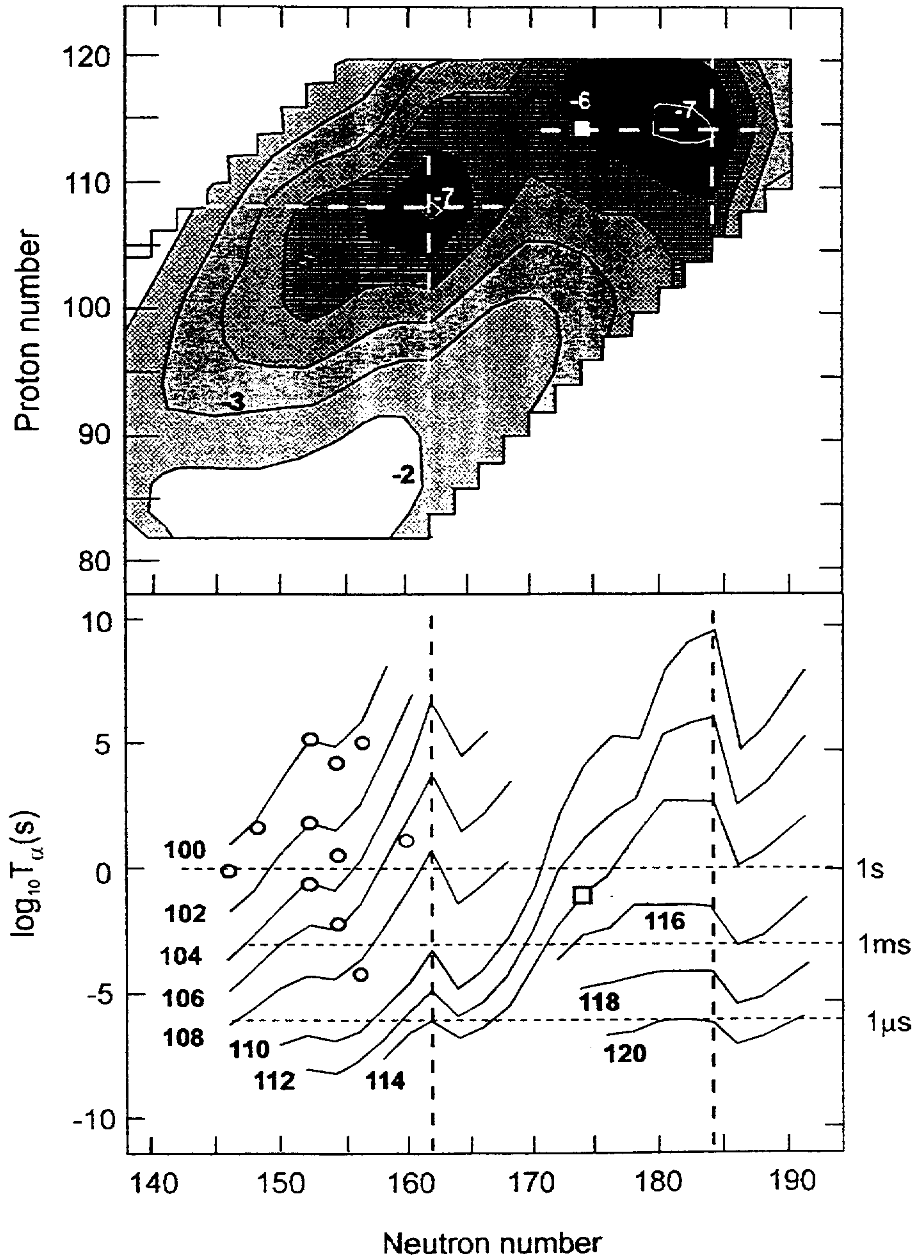


Fig.6 Contour map of the shell correction to energy. The difference in the values between neighbouring contour lines specified by the scale

Lower part: alpha-decay energy Q_α(MeV) as a function of neutron number for nuclei with Z=100-120 calculated by R.Smolanczuk et al.[25]. Experimental values for even-even isotopes are shown by open circles. Square - heaviest even-even isotope ${}^{284}114$ expected from the reaction ${}^{244}Pu({}^{48}Ca,4n)$.

spontaneous fission. It is expected that even-even nuclei in that range will undergo an α-decay with a half-life, strongly depending on the neutron excess. (Fig.6).

Definitely, the effect of a sperical shell on radioactive properties of super heavy nuclei, as seen in Fig.6, will be observed in the decay of isotopes with Z=114, 116 already for N≥174.

For the synthesis of these nuclides in the fusion reactions it is necessary to choose nuclei-partners with the maximum neutron excess. The reaction ^{244}Pu(^{48}Ca,xn)$^{292-x}$114 fulfills the requirements best of all.

In the reaction ^{244}Pu+HI the replacement of ^{34}S ions by ^{48}Ca, all other factors being equal, should lead to a further reduction of the evaporation residue formation cross section, which is already small. However, owing to a considerable mass defect in the double magic nucleus ^{48}Ca, the excitation energy of the compound nucleus ^{292}114 will be about 30 MeV and its transition to the ground state will be accompanied by a 3 or 4 neutron emission. This should increase the EVR-s formation cross section.

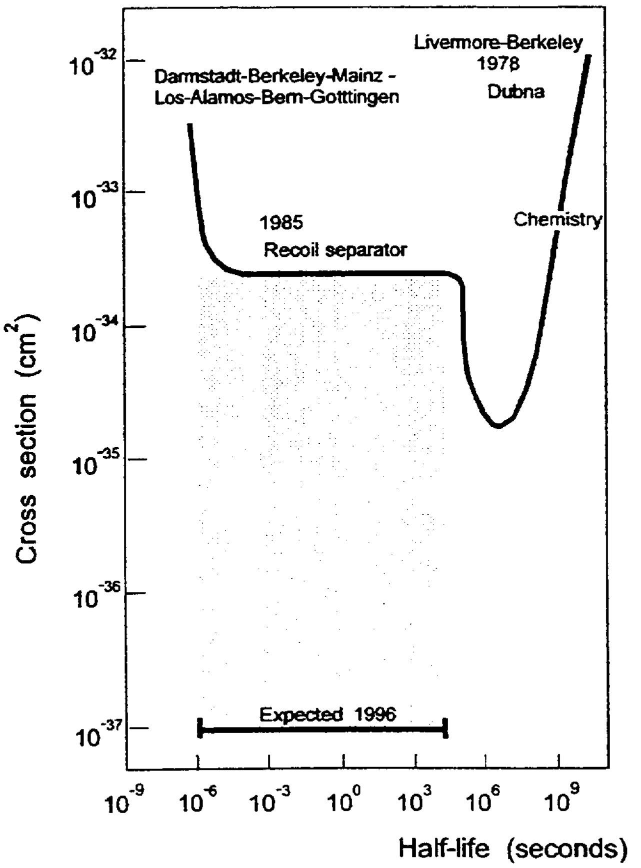


Fig.7 Cross section limit in the attempts to produce super heavy nuclei Z=116 by fusion ^{248}Cm+^{48}Ca (upper solid line). Lower line-limiting cross section expected in the ^{244}Pu(^{48}Ca,xn)$^{292-x}$114 reaction.

Such considerable variations make it inpossible to estimate accurately the expected formation cross sections of superheavy nuclei in xn-channels. It is obvious though that this cross section will be small, and the luminosity of the experiment should be high.

Development of accelerators and experimental technique does allow at present to achieve cross sections of tens of picobarns. This is almost by 3 orders of magnitude lower than in the previous attempts of synthesizing superheavy elements in the reaction ^{248}Cm+^{48}Ca (Fig.7).

We intend to perform this experiment in 1996 and have begun its preparation. An essential point of the programme is the development of a new powerful ion source ECR-4M (GANIL - DUBNA) and of an external beam injection system at the U-400 accelerator for obtaining intensive ion beams of ^{48}Ca.

We also realize that this experiment will be performed by a collaboration of scientists who are involved in the problem of synthesis and investigation of new elements.

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