Analysis of laboratory nucleosynthesis products

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We present the results of the experimental study on synthesis of a wide range of isotopes in a superdense plasma. The initial conditions necessary for plasma bunch formation were provided by specially organized coherent impact on a solid target with a total energy up to 1 kJ. More than 4000 shots were performed with various targets made of light, medium, and heavy elements. Subsequent analysis of the products of the target explosion reveals the presence of a wide range of elements absent in the initial materials. Elements with nuclei three and more times heavier than the nucleus of the target main element are detected in the products. The isotopic composition of the produced elements significantly differs from the natural one. The presence of unknown superheavy elements at the border of the periodic table and beyond it was detected by several different spectroscopic methods of elemental and isotopic analyzes.

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Introduction

This work summarizes the results of analysis of the nuclear transmutation products in the supercompressed substance obtained in 1999–2003 by the Electrodynamics Laboratory 'Proton–21' (Ukraine, Kiev) that was established to develop a novel approach for radioactive waste utilization.

The project was based on the idea that it is possible to create a special kind of disturbance impact on a solid substance that will induce, under certain conditions, the self-organizing avalanche process of supercompression up to the collapse state at which the conditions for collective multiparticle transmutation reactions of radioactive elements into stable ones will become possible, owing to the redistribution of neutrons in the substance volume and increase of their concentration at the center of the collapse. A few experimental setups have been built in the laboratory. The first successful experiment was performed on February 24, 2000. By March, 2003, 4037 dynamic compressions of solid targets were performed leading to specific explosions and the radial dispersion of a transformed target material from the collapse zone.

I. MULTINUCLEAR REACTIONS IN A SUPERDENSE SUBSTANCE

A. Experiment

The main experimental setup realizes the consecutive space—time energy compression and is capable to impact on the solid target up to 1 kJ of energy during the pulse time less than 100 ns. The electron beam is used as an

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initial carrier of the concentrated energy. On the final stage, the substance of target is compressed up to a density of $> 10^{26}\,\mathrm{cm}^{-3}$, while the estimated power density in the collapse volume exceeds $10^{22}\,\mathrm{W\,cm}^{-3}$.

The target explodes as a result of dynamic compression. At explosion an ultrabright quasipoint X-ray source is formed with registered energy range from $2\dots3\,\mathrm{keV}$ up to $10\,\mathrm{MeV}$ and more, with maximum intensity in the range $10\dots30\,\mathrm{keV}$. Products of a target explosion precipitate on the internal walls of concentrator and on the special accumulating screen — metallic disk 1 cm in diameter. The precipitated products have the form of irregularly scattered drops, beads, films, etc.

B. Analysis

Precipitated products were analyzed to determine their isotopic and elemental compositions. The analytical methods employed were:

- electron probe microanalysis (EPMA);
- Auger-electron spectroscopy (AES);
- secondary ion mass–spectrometry (SIMS);
- laser mass–spectrometry (LMS);
- Rutherford backscattering of accelerated alphaparticles (RBS).

As a rule, after the experiment, elemental analyzes of the surface and surface layer up to $3\,\mu\mathrm{m}$ deep of each accumulating screen (made of chemically pure material: Cu (99.99% pure), Ag (99.99%), Ta (99.97%), Pb (99.75%), etc.) were performed. Minimal number of microprobes was from 10 to 20, some regions (spots) were also analyzed by several methods.

By March 12, 2003, altogether 14520 analyzes were performed and registered. The summary of the analytical methods and number of analyzes are listed in Table I.

TABLE I: List of analytical methods and number of analyzes.

| Method | Number of | Number of |
|-------------------|-----------|-----------|
| | samples | probes |
| EPMA | 541 | 9231 |
| LMS | 20 | 297 |
| AES | 25 | 474 |
| SIMS | 24 | 399 |
| RBS | 40 | 40 |
| EPMA+LMS | 38 | 1227 |
| EPMA+AES | 44 | 1522 |
| EPMA+SIMS | 21 | 619 |
| EPMA+LMS+AES | 4 | 164 |
| EPMA+LMS+SIMS | 2 | 57 |
| EPMA+AES+SIMS | 7 | 316 |
| EPMA+LMS+AES+SIMS | 1 | 43 |
| LMS+AES | 1 | 29 |
| AES+SIMS | 2 | 102 |
| total EPMA | 658 | 11578 |
| total LMS | 66 | 1001 |
| total AES | 84 | 1359 |
| total SIMS | 57 | 542 |
| total RBS | 40 | 40 |
| TOTAL | 770 | 14520 |

C. Main results of elemental composition analyzes after one of the typical experiments

Analyzes show that the products of impact explosion precipitated on the accumulating screen contain a wide spectrum of the light, medium and heavy chemical elements that are absent in comparative quantities or concentrations in the initial materials involved in the process of nuclear transformation.

This fact is illustrated in Table II, where the quantities of impurities in chemically pure copper (used for the target and accumulating screen) are compared with those detected in the products precipitated on the accumulating screen after one of the typical experiments. Correlation ratios between the concentration of impurities in the target material and their quantities in the precipitated products have near zero or negative values.

The composition of the initial environment and target materials were measured with a mass–spectrometer of glow discharge VG 9000 (Thermo Elemental), with sensitivity from 100% to ppt level in a single analysis.

Three different instruments were used for analysis of the elemental composition of the accumulating screen surface layer after the target impact explosion:

• Auger spectroscope JAMP-10S (JEOL, Japan) — 21 spots $<1.0\,\mu\mathrm{m}$ in diameter and $0.002\,\mu\mathrm{m}$ deep were analyzed;

- EPMA analyzer REMMA 102 (Ukraine) 113 areas $11 \times 11 \,\mu\mathrm{m}$ and $3.0 \,\mu\mathrm{m}$ deep, and 417 extrinsic beads of different shape on the surface of the accumulating screen.
- SIMS analyzer IMS 4f (CAMECA, France) 5 areas $250 \times 250 \,\mu\text{m}$ and $0.4 \,\mu\text{m}$ deep (because SIMS is the destructive method, it was used after Auger and EPMA);

As can be seen from Table II, both the light elements (with the mass number less than the initial copper) and the heavy ones are present. It should be emphasized that the elements with atomic masses exceeding two masses of the initial element (elements with mass number Z > 58) were detected as well. Quantities of the mentioned elements by a few orders of magnitude exceed impurities in the initial material.

To get quantitative estimations for the number and the concentration of deposited atoms, the statistical analysis was performed for the following layers:

- 1. Thin surface layer about $0.002 \,\mu\mathrm{m}$;
- 2. Intermediate layer from $0.002 \,\mu\mathrm{m}$ to $0.4 \,\mu\mathrm{m}$;
- 3. Rest of the material layer from $0.4 \,\mu\mathrm{m}$ to $3.0 \,\mu\mathrm{m}$ (maximum depth of analyzes).

As the result of statistical evaluation, the concentrations in the above–mentioned layers were obtained for each element. The validation criterion was the accordance between the calculated element concentration and concentration measured by all applied instruments.

Statistical analysis of the quantity of deposited atoms revealed the following:

- Deposited atoms $(2.8 \times 10^{16} \text{ atoms})$ concentration is maximum in the surface layer $0.002 \,\mu\text{m}$ thick (1st layer) and the concentration of copper (main target and screen material) is lower than $10 \,\%$.
- Main quantity of deposited elements (1.3 \times 10¹⁸ atoms) and the majority of the elements with masses exceeding two masses of the initial material (i.e. Cu) are present in the 2nd layer (From 0.002 to 0.25 μ m).
- Quantity of deposited atoms in the 3rd layer $(0.25\,\mu\mathrm{m})$ and down to the limit of the analyzer, $3.0\,\mu\mathrm{m})$ is 3.1×10^{17} atoms.

Total number of deposited atoms, excluding copper, present in the substance up to $3.0\,\mu\mathrm{m}$ deep was calculated as sum of the atom quantities in the volumes of each layer and equals 1.6×10^{18} atoms. Change of the thickness of the 2nd layer containing the main part of deposited elements from $0.2\,\mu\mathrm{m}$ to $0.4\,\mu\mathrm{m}$ gives the range $(1.4\dots2.4)\times10^{18}$ for the total number of deposited atoms.

TABLE II: Number of atoms in the surface layer of accumulating screen.

| Element | Z | Init Cu target | Accum. screen |
|---------------------|----|----------------------|----------------------|
| Li | 3 | $1.7\mathrm{E}{+}12$ | $6.0\mathrm{E}{+}11$ |
| $_{\mathrm{Be}}$ | 4 | $6.1\mathrm{E}{+}11$ | $1.3\mathrm{E}{+}14$ |
| В | 5 | $2.1\mathrm{E}{+}12$ | $4.1\mathrm{E}{+}13$ |
| $^{\mathrm{C}}$ | 6 | _ | $9.5\mathrm{E}{+}17$ |
| N | 7 | _ | $1.1\mathrm{E}{+}15$ |
| O | 8 | _ | $4.3\mathrm{E}{+}15$ |
| Na | 11 | $6.5\mathrm{E}{+}13$ | $1.3\mathrm{E}{+}16$ |
| Mg | 12 | $3.6\mathrm{E}{+}13$ | $3.3\mathrm{E}{+}15$ |
| Al | 13 | $3.9\mathrm{E}{+}14$ | $3.3\mathrm{E}{+}17$ |
| Si | 14 | $3.8\mathrm{E}{+}13$ | $9.8\mathrm{E}{+}16$ |
| P | 15 | $6.5\mathrm{E}{+}14$ | $2.0\mathrm{E}{+}16$ |
| \mathbf{S} | 16 | $3.4\mathrm{E}{+}14$ | $1.2\mathrm{E}{+}17$ |
| Cl | 17 | $2.4\mathrm{E}{+}10$ | $1.5\mathrm{E}{+}17$ |
| K | 19 | _ | $5.3\mathrm{E}{+}16$ |
| Ca | 20 | $3.2\mathrm{E}{+}14$ | $1.8\mathrm{E}{+}16$ |
| Ti | 22 | $2.3\mathrm{E}{+}12$ | $3.8\mathrm{E}{+}15$ |
| V | 23 | $1.1\mathrm{E}{+}11$ | $9.1\mathrm{E}{+}13$ |
| Cr | 24 | $3.3\mathrm{E}{+}12$ | $2.5\mathrm{E}{+}15$ |
| Mn | 25 | $2.4\mathrm{E}{+}13$ | $1.5\mathrm{E}{+}15$ |
| Fe | 26 | $1.3\mathrm{E}{+}15$ | $8.7\mathrm{E}{+}16$ |
| Co | 27 | $1.0\mathrm{E}{+}12$ | $3.9\mathrm{E}{+}14$ |
| Ni | 28 | $3.8\mathrm{E}{+}14$ | $2.0\mathrm{E}{+}14$ |
| Zn | 30 | $5.5\mathrm{E}{+}13$ | $7.5E{+}16$ |
| Y | 39 | $1.9\mathrm{E}{+}10$ | $2.0\mathrm{E}{+}14$ |
| Zr | 40 | $5.9\mathrm{E}{+}10$ | $2.8\mathrm{E}{+}13$ |
| Ag | 47 | $8.5\mathrm{E}{+}13$ | $6.4\mathrm{E}{+}15$ |
| Cd | 48 | $1.1\mathrm{E}{+}12$ | $2.2\mathrm{E}{+}15$ |
| In | 49 | $9.7\mathrm{E}{+}11$ | $1.9\mathrm{E}{+}15$ |
| Sn | 50 | $2.0\mathrm{E}{+}13$ | $1.6\mathrm{E}{+}16$ |
| Te | 52 | $8.6\mathrm{E}{+}12$ | $1.4\mathrm{E}{+}15$ |
| $_{\mathrm{Ba}}$ | 56 | $3.2\mathrm{E}{+}11$ | $2.4\mathrm{E}{+}15$ |
| La | 57 | $1.4\mathrm{E}{+}10$ | $7.2E{+}14$ |
| Ce | 58 | $2.2\mathrm{E}{+}10$ | $2.5\mathrm{E}{+}15$ |
| \Pr | 59 | $2.6\mathrm{E}{+}10$ | $1.5\mathrm{E}{+}14$ |
| Ta | 73 | _ | $4.2\mathrm{E}{+}15$ |
| W | 74 | $3.1\mathrm{E}{+}11$ | $2.3\mathrm{E}{+}16$ |
| Au | 79 | $1.0\mathrm{E}{+}11$ | $5.8\mathrm{E}{+}15$ |
| Pb | 82 | $2.5\mathrm{E}{+}13$ | $2.0\mathrm{E}{+}17$ |
| TOTA | L | $3.7\mathrm{E}{+}15$ | $2.2\mathrm{E}{+}18$ |

• Quantity of deposited atoms in randomly dispersed particles on the surface of the accumulating screen is equal to 6.0×10^{17} atoms.

The total number of deposited atoms, excluding copper, after the experiment in comparison to the initial concentration is shown in Table II.

The same estimation for the synthesized atoms was derived from the 'marked target' experiments. The targets for these experiments were fabricated from radioactive cobalt (60 Co). After an impact, the system activity (number of radioactive decays per second) decreased by the amount equal to the transmutation of 10^{18} atoms of the active target volume — intensity of the 60 Co spectral lines decreased while no other radioactive isotopes line did appear.

D. Isotopic composition analyzes of accumulating screens

To confirm that we deal with the nucleosynthesis process, the isotopic composition of the products on the surface of the accumulating screens was analyzed. The stimulus for this was the well–known fact that the isotopic composition of any element is almost identical throughout the Solar system (see, e.g. [1, 2]).

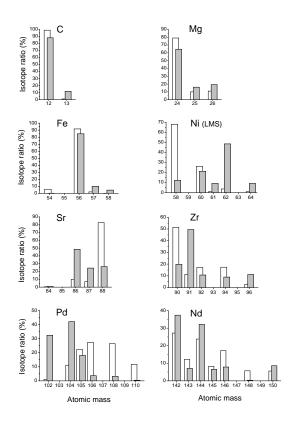


FIG. 1: Isotopic composition of some elements measured with LMS (indicated) and SIMS (others). Natural composition is depicted with empty bars, the composition of synthesized elements with hatched bars.

Analyzes of the isotopic composition were done with two different methods, i.e., LMS and SIMS (see Table I). As was found out, most of the analyzed spots of an accumulating screen had an isotopic composition that differs from a natural one. Examples of the isotopic composition for some elements are presented in Fig. 1.

As can be seen from this figure, the isotopic composition of the elements synthesized from the copper target, differs significantly from the natural one. Hence, this proves an artificial origin of the detected nuclei.

II. SYNTHESIS OF THE ELEMENTS AT THE BORDER OF THE PERIODIC TABLE AND BEYOND IT

The presence of heavy elements (tantalum, tungsten, gold, and lead) among the products of nucleosynthesis from the copper target encouraged us to carry out the experiments with heavier targets. In fact, since lead $(A(Pb) \approx 207)$ was produced from the copper target $(A(Cu) \approx 64)$, one should expect in similar way to get elements at the border of the periodic table and even beyond it if a target made from heavier elements is used.

For this aim, the experiments with the platinum, lead, and bismuth targets were carried out. The results of analyzes revealed the presence of long-living isotopes of chemical elements at the border and even beyond the known part of periodic table.

To identify the elemental composition of the accumulating screen surface, the Auger spectroscopy (AES) method was used. The method is non-destructive and highly localized $(50...100\,\mathrm{nm})$. The depth of analyzes is low $(1...2\,\mathrm{nm})$. The method ensures a wide range of detectable elements (all but H and He) and a relatively high sensitivity $(0.1...1\,\mathrm{at.\%})$.

The spectra obtained with the help of a Augermicroprobe JAMP-10S (JEOL, Japan) typically contained Auger peaks of a considerable amount of chemical elements that hindered their identification. To identify the Auger peaks, the measurement was performed in a wide range of energies (30...3000 eV, energy resolution 0.5...1.2%) to cover the maximum number of Auger peaks' series, and prolonged exposures (up to 3 hours) were used to identify the low intensity peaks. The spectra were recorded in differential form at the accelerating voltage of the electron probe of 10 kV and beam current $10^{-6} \dots 10^{-8}$ A. Residual pressure in the sample chamber was 5×10^{-7} Pa. Artifacts like electrical charging, characteristic energy losses, and chemical shift were taken into consideration. A standard concentration calculation program supplied by the manufacturer (JEOL) was used for quantitative analysis.

In the process of analysis, the unidentified peaks with energies of 172, 527, 1096 eV, and a doublet of 130 and 115 eV were registered on the surface of accumulating screens. These peaks do not correspond to any of the catalogued peaks of chemical elements and cannot be referred to any of the known artifacts. One of the possible explanation of the mentioned peaks is the presence of long-living transuranium elements.

Two sections of the Auger spectrum (energies 172, 527 eV) out of six unidentified peaks are presented in Fig. 2.

To provide elemental analysis of the accumulating screens surface in a wide range of masses, a SIMS analyzer IMS 4f (CAMECA, France) was used.

Here are some key characteristics of the instrument:

• mass detection range — up to 480;

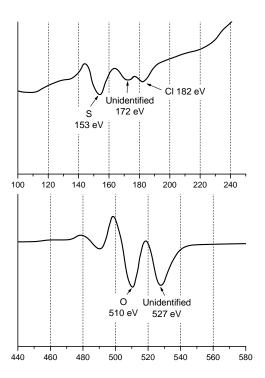


FIG. 2: Sections of Auger spectra with unidentified peaks.

- mass resolution $(M/\Delta M)$ 2500;
- locality $0.2 \,\mu\text{m}$;
- primary ions: Cs^+ , O_2^+ , O^- , Ar^+ ;
- secondary beam stability (20 min): $\Delta I/I = 0.7 \%$, $\Delta M/M = 7 \times 10^{-6}$.

A complex approach was used for elemental analyzes of the accumulating screens surface and included studies of the topography of chemical elements on the surface in the cluster suppression mode (Offset) and high mass resolution mode simultaneously with the photo film recording of the mass distribution.

In the Offset mode, an additional voltage is applied and, owing to the potential barrier, allows us to identify monoatomic ions even in the low–mass resolution mode and, hence, to exclude clusters and multicharged ions.

From the whole mass—spectrum, we consider as unidentified only those peaks which (a) were not observed in the reference material spectrum, (b) did not coincide with the other elements' distribution on the screen surface, (c) offset voltage increase led to an increase of the intensity in comparison with clusters' peaks.

Typical mass spectrum in the mass range 250...280 obtained by IMS 4f is presented in Fig. 3. Filled peaks, corresponding to 265 and 267, are unidentified; 268, 270, 272, 274 are Cu₄O. Increasing the offset voltage led to a significant decrease of the intensity for copper clusters

and the negligible fall of the intensity of 265 and 267 peaks indicating that they belong to monoatomic ions. To exclude possible chemical elements combinations, the photos of all present elements distributions were done, and they show that none of the unidentified masses coincide with any of them.

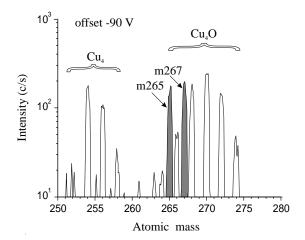


FIG. 3: Mass spectrum revealing the presence of unidentified masses.

Up to date, 20 samples were thoroughly checked for unidentified masses. On 17 samples, more than 100 unidentified masses were found in the range 221...475. Most frequently were detected the peaks of 271, 272, 330, 341, 343 masses. Accumulating screens that were used repeatedly in experiments with copper targets also revealed the presence of long-living isotopes in the mass range 300...481.

In addition to above—mentioned spectrometric methods, the samples were examined for the presence of superheavy elements with the most direct method — Rutherford backscattering. The samples were irradiated with the beam of 27.2 MeV alpha—particles accelerated in a U–120 cyclotron [4]. The energy spectrum of scattered projectiles exhibited the scattering centers corresponding to mass numbers in the range $200 \dots 400$ (Fig. 4).

Thus, different methods of analysis detected the presence of unidentified superheavy elements among the products of artificial nucleosynthesis.

III. SUMMARY OF RESULTS AND DISCUSSION

We have presented the survey of experimental studies of nucleosynthesis products obtained in experiments with the supercompressed substance. The essence of results is as follows:

• In substance compressed up to a superhigh density (presumably, $10^{26} \,\mathrm{cm}^{-3}$), the process of nucleosynthesis and transmutation occurs. This process

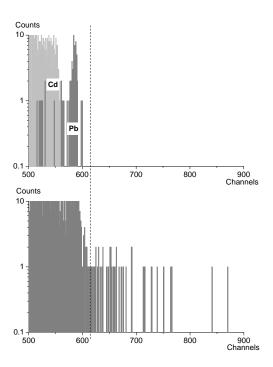


FIG. 4: Rutherford backscattering data; initial material (above) and after the impact (below).

takes place over a macrovolume of the target substance $(2...5 \times 10^{18} \text{ atoms/kJ})$.

- Nuclear processes in a target are collective and multiparticle that is proved by significant amount (> 10¹³) of synthesized nuclei with masses more than two masses of the initial nucleus.
- No α-, β-, γ-active isotopes were observed in the products of laboratory nucleosynthesis, the radiation intensity never exceeded the background intensity.
- The activity of the targets marked with radioactive isotopes was reduced after compression impact by the value equal to the transmutation of $\sim 10^{18}$ atoms of the active target zone (collapse zone) for every 1 kJ of the driver energy.
- Developed installation has high reproducibility in reaching the conditions in a compressed substance necessary for the ignition of collective multiparticle fusion–fission reactions of the full spectrum of chemical elements.
- The products of nuclear transmutation reveal the presence of long-living isotopes of superheavy elements.

We consider that these reactions are, in some sense, similar to the pycnonuclear reactions that take place under the specific conditions of compact astrophysical objects (white dwarfs, supernovas, outer shells of neutron stars, etc.).

The new challenge is the development of a method and a technique for simultaneous measurement of both the atomic and nuclear properties of superheavy atoms, i.e., nucleus charge and mass.

The results of our experiments allow us to suppose that the synthesized nuclei are the product of clusterization in the decaying superdense electron–nucleon plasma that corresponds to maximization of the binding energy per nucleon, dependent on the substance density, on the one hand, and on the neutron concentration in the certain volume of clusterization, on the other hand.

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