STUDY OF THE COMPOSITION OF PRODUCTS OF CONTROLLED NUCLEOSYNTHESIS BY LOCAL AUGER-ELECTRON SPECTROSCOPY

S. V. Adamenko, A. S. Adamenko, and S. S. Ponomarev^{*}

Electrodynamics Laboratory "Proton–21" 14/1 Dovjenko Str., 03057, Kiyv, Ukraine[†]

(Dated: April 27, 2004)

By local Auger-electron spectroscopy on solid targets and accumulating screens, we studied the composition of nucleosynthesis products, in which we expected to reveal the presence of long-lived transuranium elements (LTE). The goal of the work was to find LTE on the surface of the specimens under investigation. The specificity of the surface composition determination consisted in that the registered spectra included usually the Auger-peaks of a lot of chemical elements, which poses major difficulties in their identification. To solve the latter problem, we used a wide range of energies (from 30 to 3000 eV) with the purpose to cover a maximum number of series of the Auger-peaks of analyzed elements and long exposures (up to 3h) to reveal Auger-peaks with small intensity. In a number of cases for analyzed elements in complicated spectra the Auger-spectra of corresponding pure elements or their simple compounds were registered with a high signal-to-noise ratio. As artifacts of the analysis, we consider such phenomena as the electric charging, characteristic losses of energy, and chemical shift. On the study of the surface of specimens, we found the unidentifiable Auger-peaks with energies of 172, 527, 1096, 94, and 560 eV and the doublet of peaks with energies of 130 and 115 eV. We failed to refer them to any Auger-peaks of chemical elements in the atlases and catalogs or to any artifacts. As one of the variants of interpretation of the revealed peaks, we consider the assumption about their affiliation to LTE.

Keywords: controlled nucleosynthesis, composition of nucleosynthesis products, Auger-electron spectroscopy, unidentifiable Auger-peaks, long-lived transuranium elements

INTRODUCTION

In the present work, we study the specimens whose surfaces contain the products of the explosion-induced dispersion of targets undergone the high-energy impact compression which was realized with the purpose to reach the extremal density of a substance and, presumably, to initiate the collective processes of nuclear transformations (nucleosynthesis) terminating in the formation of highly stable nuclei (stable and long-lived isotopes) of various chemical elements with broad mass spectrum including transuranium ones [1, 2].

As specimens, we take the solid targets, in which the reaction of nucleosynthesis ran, and accumulating screens, on which the target material undergone a nuclear transmutation is deposited after its explosion. The products remained in the target crater and deposited on accumulating screens are drops, splashes, films, particles, and other micro- and nanoobjects with complicated morphology which are irregularly distributed on their surfaces. In the study of the reaction products, we were interested, first of all, in the determination of their element and isotope compositions. It is especially worth to note that we expect to reveal the presence of LTE.

The isotope composition of the nucleosynthesis products was carried out by laser mass-spectrometry (LMS)

[†]Electronic address: enr30@enran.com.ua

and secondary ion mass-spectrometry (SIMS). The results of these investigations are far beyond the frame of the present work and now are prepared as separate publications. As for the problem of determination of their element composition, we used the complex of mutually supplementing physical methods rather than a sole one. On the one hand, these methods must have high sensitivity and, on the other hand, must cover the range of typical sizes of all analyzed objects (from several mm to several nm). In fact, the analyzed chemical elements, in particular LTE, can be distributed uniformly in the object under study with low concentration in it or, on the contrary, be concentrated in small particles and/or thin submicron films on the specimen surface and on the inner interfaces between elements of the structure which compose the surface layer. It is also clear that the employed methods must overlap the range of thicknesses of the studied layers since the analyzed elements can be distributed irregularly throughout depth.

We chose local Auger-electron spectroscopy (AES) as the method for quantitative determination of the composition of microobjects (the objects with size more than $1 \mu m$ were studied by X-ray electron probe microanalysis) which were contained in the nucleosynthesis products and had at least one submicron size (the surface proper, inner interfaces, submicron particles and films). Such a choice was stipulated by that the method well satisfies the requirements given above. First of all, AES is a nondestructive method for quantitative determination of the composition, and the specimens being studied with it can be investigated with other methods. The method possesses a high spatial locality (100...50 nm), a small

^{*}Institute of Problems of Materials Science of the NAS of Ukraine, 3 Krzhizhanovsky Str., Kyiv, 03142 Ukraine

depth of the analyzed region (1...2 nm), a wide range of registered elements (all except for H and He), and a quite high sensitivity (0.1...1 at. %)). If one uses the method of ion etching, AES also allows one to study the distribution of elements throughout depth [3–9].

Thus, the goal of the present work is to study the element composition of the nucleosynthesis products by AES and to establish the presence or the absence of LTE.

I. MATERIALS AND METHODS

We determined the composition of the specimen surface with an Auger-microprobe JAMP-10S (JEOL, Japan). Spectra were registered at the accelerating voltage of the electron probe which was equal to 10 kV, a beam current of $10^{-6} \dots 10^{-8}$ A, and the residual pressure of 5×10^{-7} Pa in a specimen chamber. By considering such an artifact as characteristic energy losses, we used the accelerating voltages of 5 and 3 kV. The energy range of a semicylindical mirror energy-analyzer of the Auger-spectrometer was 30...3000 eV and its energy resolution was from 0.5 % to 1.2 %. All spectra were registered in differential form. In the quantitative analysis, we used the standard program for the computation of the concentrations of elements given by the JEOL firm-producer.

As the objects of our investigation, we took the targets which were made of light, medium, and heavy chemically pure metals with atomic masses from 9 to 209, as well as the accumulating screens made of chemically pure Cu, Ag, Ta, Au, and Pb. The screens were disks from 0.5 to 1 mm in thickness and from 10 to 15 mm in diameter and served as a substrate. On one of the screen surfaces, the studied layer of a material consisting of the nucleosynthesis products was located. The layer had a slightly pronounced relief and the axial symmetry. Its area varied from 1 to 2 cm^2 . In the determination of the surface composition, we used as-received specimens, i.e., the analyzed surface was not undergone to any cleaning procedure.

II. RESULTS AND DISCUSSION

Up to now, we have studied more than 100 specimens by AES. In all the cases (including those where the target and an accumulating screen were made of one chemical element maximally purified from impurities), we registered from twenty to thirty chemical elements in specimens. The amounts of these elements exceeded the total content of impurities in the initial materials by several orders of magnitude. As usual, the specimens under study contained mainly chemical elements with small and medium atomic numbers. The fraction of heavy chemical elements is lower, but it increases with the atomic number of a target. Experimental data derived by several methods (including AES) are presented in [1] by the example of

the comparative analysis of the registered composition of nucleosynthesis products on a target and an accumulating screen made of pure copper. At present, we have a huge database (about 800 specimens and 14500 analyses) on the composition of the products of laboratory nucleosynthesis which was obtained by the methods of AES, SIMS, X-ray electron probe microanalysis, etc. It is obvious that there is no point in giving one or several lists of the results of analysis of the nucleosynthesis products derived by AES. Moreover, the analysis of this database and the construction of correlation dependencies between the compositions of products, targets, accumulating screens and the parameters of the technological process go beyond the frame of this work. Below, we will not concentrate on the presence of well-known chemical elements in the analyzed specimens and will pay attention to the attempt to answer the question about the content of LTE in the nucleosynthesis products.

The specificity of determination of the composition of a material layer surface under study was defined by the fact that the registered Auger-spectra contained, as usual, the Auger-peaks of at least 10 chemical elements. Moreover, the collection of these elements can considerably vary not only from one specimen to another but from one analysis point to another in the limits of the same specimen, and the content of elements changed in a wide range. This yields that the spectra contain a great number of peaks which frequently overlap one another. This circumstance hampers the identification of the registered Auger-peaks.

There was a number of other factors concerning the problem of identification of the registered Auger-peaks. As usual, we registered Auger-spectra in the energy range 30... 3000 eV. The choice of such a wide range was caused by two reasons. On the one hand, this choice allows us to cover the maximum number of series of the Augerpeaks of analyzed elements and must promote solving the problem of their identification in the low-energy region. On the other hand, it is necessary to answer the question about the presence of LTE in specimens, because LTE being heavy have a lot of peaks in the highenergy region. However, the known atlases and catalogs of Auger-electron spectra (e.g., [10–12]) usually used in the identification of registered Auger-peaks contain the extremely limited volume of data for the energy range 1000...2000 eV and no data for the range $2000...3000 \,\mathrm{eV}.$

Discussing the problem of identification of Augerpeaks, we should like to indicate one more circumstance. The sensitivity of the method of Auger-electron spectroscopy in the determination of the amount of heavy elements is less than that for light elements [3–5]. Therefore, with the purpose to reveal small amounts of the required elements and to enhance the signal-to-noise ratio in the registered spectra, we used long exposures (up to 3 h) and large currents of the primary beam of electrons (up to 10^{-6} A). However, the reference catalogs of Auger-electron spectra [10–12] which are used upon the identification of Auger-peaks give a clear preference to high energy resolution (the registration was realized on narrow slits of spectrometers) rather than to large signalto-noise ratio. In other words, Auger-peaks with low intensity were sacrificed by the reference literature to the data on a fine structure of analytical Auger-peaks.

Therefore, as a result of the above-mentioned circumstances, a number of Auger-peaks absent in the reference catalogs were registered by us in the specimens under study. The identification of each of these peaks was carried out according to the following scheme. For the Auger-spectrum where a unidentifiable peak was observed, we determined the collection of elements whose Auger-peaks were present in such an Auger-spectrum. For each of these elements, we registered its reference Auger-spectrum in the energy range 30...3000 eV with high signal-to-noise ratio (the large duration of the exposure) with the purpose to reveal the low-intensity Auger-peaks of the chosen element. The reference Augerspectra were registered on specimens made of relevant pure elements or their simple compounds. Then we made attempt to refer the unidentifiable peak to some lowintensity Auger-peak revealed in the reference Augerspectra of the chemical elements present in the analyzed spectrum. In such a way, we identified, for example, the low-intensity Auger-peaks of series KLL of Si and Al with energies, respectively, of 1737 and 1485 eV.

Some unidentifiable peaks were referred to artifacts of the analysis. As artifacts, we considered such phenomena as the electric charging, characteristic losses of energy, and chemical shift. The criterion for the unidentifiable peak to be considered as a peak of characteristic losses of energy was its shift or disappearance upon a change in the accelerating voltage of the primary electron beam. The criterion of appearance of the unidentifiable peaks as a result of the electric charging was unproper energetic position of the Auger-peaks of chemical elements contained in the analyzed Auger-spectrum and the lack of reproducibility of its energetic position upon a change of high voltage and current of primary electron beam. While analyzing a chemical shift, we took into account, first of all, its value and the chemical environment of the analyzed peak.

After the implementation of all the above-presented stages of the analysis, we registered 6 unidentifiable peaks. This amount was accumulated to the time of writing the present work after the study of the composition of surface layers of more than 100 specimens. The brief information about them is systematized in Table I.

The unidentifiable peak with an energy of 172 eV was registered on 5 specimens. It is contained in 11 Augerspectra registered on these specimens. A fragment of one of these Auger-spectra containing the unidentifiable peak with an energy of 172 eV is shown in Fig. 1. The number of such spectra, by one's wish, can be increased to any value, because the part of the surface of each specimen where this peak can be registered is rather extended. The peak was registered on the metal matrix surface. Its intensity is small in all the cases and decreases from the

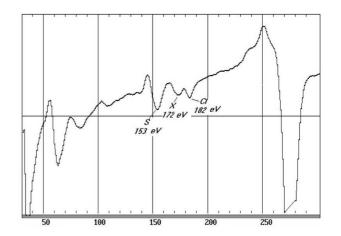


FIG. 1: Fragment of a typical Auger-spectrum containing the unidentifiable peak with an energy of 172 eV.

specimen center to its periphery. The energy position of the peak is reproduced quite exactly. Its typical chemical environment is Si, S, Cl, K, C, Ca, N, O, Cu, Zn, and Na, whereas nonmetallic elements except for C and O are present usually in slight amounts.

We say a few words about the stability of the peak. After the registration of the Auger-spectrum, if we at once detect it once more on the same place, the peak intensity decreases usually by $20 \dots 30\%$, whereas the repeated subsequent registrations reveal no decrease in its intensity. We note that, in this case, no significant change in the intensity of other peaks, in particular of the Auger-peaks of C and O, occurs. That is, the mentioned phenomenon cannot be referred to the presence of a carbon deposit and looks as if the substrate of this peak is decomposed in the subsurface layers under the action of the electron beam of the probe, where this action is most intense, and remains stable at large depths because of the decrease in the level of action of the beam. The intensity of this peak is also decreased in time, which is demonstrated by Fig. 2. We present 2 Auger-spectra which contain the unidentifiable peak with an energy of 172 eV and were registered on the same place in 1 month one after another: (a) and (b), respectively. In the later case (b), the peak intensity is clearly less than that in case (a).

It is necessary to indicate that the unidentifiable peak with an energy of 172 eV appears always against the background of the Auger-peaks of S and Cl with energies of 152 and 181 eV, respectively. Nevertheless, we do not refer the mentioned peak to such an artifact of the analysis as chemical shift. Indeed, the Auger-peaks of S and Cl are present, on the one hand, in spectra in all the cases. On the other hand, even if we interpreted the unidentifiable peak as the chemically shifted Augerpeak of Cl, this shift would have an unlikely huge value (see Table II, where we give the data on most significant known chemical shifts of the Auger-peaks of S and Cl in their compounds).

Peak energy, eV	Typical element environment	Registration place	Behavior in time and under a probe	Specimen	Number of obser- vations
172	Si, K, Ca, Na, Cu, Zn, O, N, C, S, Cl	matrix	relatively stable	$\begin{array}{c} 132,137,\\ 4961,5239,\\ 5292 \end{array}$	11
1095-1098	Si, K, Ca, Na, Cu, Zn, O, N, C, S, Cl	matrix	stable	$132, 137, \\4961, 5239, \\5292$	11
525-528	Si, Al, Ca, O, C, S, Cl, Cu, Zn	particles	Ca grows in time	4169, 4540	3
130 (115)	Al, O, C, N, S, P, Cl, Cu, Sn, Ce	matrix	the intensity decreases	5633	2
94	C, O, Cu	particles at about $1 \mu m$ depth	stable	6215, shell fragment	10
560	C, O, Cu	particles	the intensity decreases	6754	1

TABLE I: Data on unidentifiable peaks in the registered Auger electron spectra.

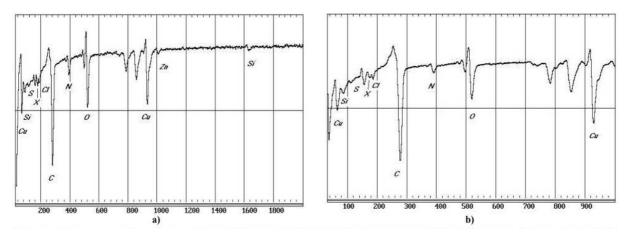


FIG. 2: Typical Auger-spectrum containing the unidentifiable peak with an energy of $172 \,\mathrm{eV}$ (a) and the spectrum registered on the same place in a month (b).

TABLE II: Data on the chemical shifts of the Auger-peaks of S and Cl in their compounds [12].

	Com- pound		$\begin{array}{c} \Delta E(\mathrm{Cl})_{\mathrm{LVV}},\\ \mathrm{eV} \end{array}$	Com- pound		$\begin{array}{c} \Delta E(\mathrm{S})_{\mathrm{LVV}},\\ \mathrm{eV} \end{array}$
ĺ	LiCl	179	-2	PbS	149	-3
	NaCl	182	1	Ag_2S	148	-4
	CuCl_2	178	-3	US_x	147.5	-4.5
	KCl	178	-3			

Chronologically, the unidentifiable peak with an energy of 172 eV was the first to be observed. Because we expected the presence of LTE in the specimens and the main goal consisted in their finding, we assumed that the detected peak could be one of the brightest characteristic Auger-peaks of some LTE which is present at the analyzed point of the specimen surface in a small amount. Then the Auger-spectrum could contain other Auger-peaks of some series of the same LTE with lower intensity. In other words, it was necessary to thoroughly test the entire energy range of the Auger-spectrum under consideration for the presence of low-intensity peaks.

The "first-shot" registration of Auger-spectra with large exposure duration on those places of the specimen surface, where the unidentifiable peak with an energy of 172 eV was found, showed a new unidentifiable peak with an energy of about 1096 eV. A high-energy fragment of one of these Auger-spectra is presented in Fig. 3. We emphasize that the new peak was registered in all cases, without exception, where the 172-eV peak was observed (see Table I), i.e., they are observed as a couple. By virtue of the weak intensity of the 1096-eV peak, its exact energy position cannot be point out due to the background fluctuations. Approximately, we may say about the interval from 1095 to 1098 eV. The same reasons hamper the study of its behavior in time or under the action of the electron beam of the probe, but it is clear that the

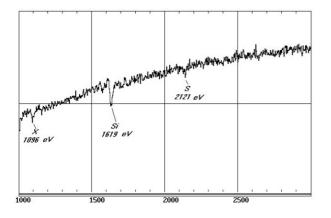


FIG. 3: High-energy part of the Auger electron spectrum, in the low-energy part of which the unidentifiable peak with an energy of $172 \,\mathrm{eV}$ is present; the exposure duration was 3 h. The spectrum contains the unidentifiable peak with an energy of $1096 \,\mathrm{eV}$ which regularly accompanies that with an energy of $172 \,\mathrm{eV}$.

The next unidentifiable peak has energy in the the interval from 525 to 528 eV. A fragment of the Auger-spectrum containing this peak is given in Fig. 4. This peak was registered in 3 Auger-spectra on 2 specimens (see Table I). In all the cases, it was found on the surface of globular particles of the second phase (inclusions) of $50...70 \,\mu$ m in diameter. Its intensity varied from a low to considerable one (see Fig. 4). The peak appeared in the typical chemical environment: Si, S, Cl, C, Ca, O, Cu, Zn, and Al. The data on the chemical shift of the Auger-peak of O are presented in Table III.

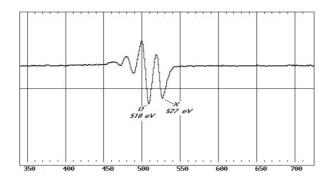


FIG. 4: Fragment of the Auger-spectrum containing the unidentifiable peak with an energy of 527 eV.

As for the stability of this 527-eV peak, we mention a few points. Its behavior under the action of the electron beam of the probe is such that, upon the repeated registration of the Auger-spectrum which was measured at once after the previous one, we failed to observe it though its intensity was significant upon the first registration. Unfortunately, the Auger-spectra illustrating this fact were observed by an operator in the scanning regime and were not registered as those of no interest at

TABLE III: Data on the chemical shift of the Auger-peak of O in its compounds [12].

_					
Compound	$\begin{array}{c} {\rm E(O)} \\ {\rm KLL}, \\ {\rm eV} \end{array}$	$\begin{array}{c} \triangle E(O) \\ KLL, \\ eV \end{array}$	Compound	E(O) KLL, eV	$\begin{array}{c} \triangle E(O) \\ KLL, \\ eV \end{array}$
Mg(OH) ₂	503	-7	BeO	510	0
$SrTiO_3$	503	-7	MgO	503	-7
α -Al ₂ O ₃	508	-2	$Al(OH)_3$	511	1
SiO_2	507	-3	$Ca(OH)_2$	511	1
$Ca(OH)_x$	512	2	CaO	509	-1
MnO_2	515.1	5.1	FeO	510	0
Fe_2O_3	508	-2	NiOOH	511	1
Ni ₂ O ₃	512	2	CuO	502	-8
Cu_2O	509	-1	Y_2O_3	507	-3
$InPO_x$	508	-2	Sb_2O_5	507	-3
Nd_2O_3	511	1	Dy_2O_3	510	0
Tb_2O_3	510	0	$\mathrm{Tm}_2\mathrm{O}_3$	505	-5
Lu_2O_3	510	0	HfO_x	510.8	0.8
PbO	513	3	$\mathrm{SiN}_x\mathrm{O}_y$	508	-2
LiNbO ₃	512	2	$BaTiO_3$	509	-1
γ -2CaOSiO ₂	506	-4	$^{\dagger}3\mathrm{CaOSiO}_2$	504	-6
β -2CaOSiO ₂	510	0	$^{\dagger}3\mathrm{CaOSiO}_2$	511	1
$CaOSiO_50.75TiO$	512	2	$^{\dagger}\mathrm{CaOSiO}_{5}0.5\mathrm{TiO}$	509	-1
$CaO2Al_2O_3$	506	-4	$^{\dagger}\mathrm{Ca_{3}Al_{2}O_{6}}$	508	-2
$^{\dagger}3CaOAl_2O_3$	513	3	$^{\dagger\dagger}3\mathrm{CaOAl}_{2}\mathrm{O}_{3}$	516	6
$12CaO7Al_2O_3$	513	3	2CaOFeO	509	-1
$4 CaOAl_2O_3Fe_2O_3$	513	3	$\mathrm{KAl}_3\mathrm{Si}_3\mathrm{O}_{12}$	502	-8

[†]After hydration.

^{††}Prior to hydration.

that moment. However, in two months, we returned to that particle, on which the intensity of the 527-eV peak was maximum, and studied again the composition of its surface. Both relevant Auger-spectra are given in Fig. 5. They also demonstrate the fact that the content of Ca has considerably increased.

As in the previous case, we tried to find the relevant low-intensity peaks for the unidentifiable 527-eV peak. However, we revealed no unidentifiable accompanying peaks upon the registration of Auger-spectra in a wide energy range and with large exposure duration on those places of specimens, where we registered the 527-eV peak.

The fourth unidentifiable peak or, more correctly, a doublet of peaks was registered only on one specimen. The main peak of the doublet had energy of 130 eV, and the energy of the less pronounced peak was 115 eV. A fragment of the Auger-spectrum containing these peaks is given in Fig. 6, a. These peaks were registered only in 2 Auger electron spectra (see Table I). In both cases, the peaks were found on small $(5 \dots 10 \,\mu\text{m}$ in diameter) light particles. The energy position of these peaks is quite exactly reproduced, but their intensity varies from a huge to insignificant one. Their typical chemical environment consists of Al, O, C, N, S, P, Cl, Cu, Sn, and Ce.

As for the stability of these peaks, we say, firstly, a few

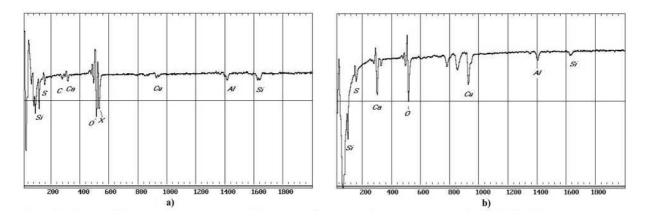


FIG. 5: Typical Auger-spectrum containing the unidentifiable peak with an energy of $527 \,\mathrm{eV}$ (a) and the spectrum registered on the same place in two months (b).

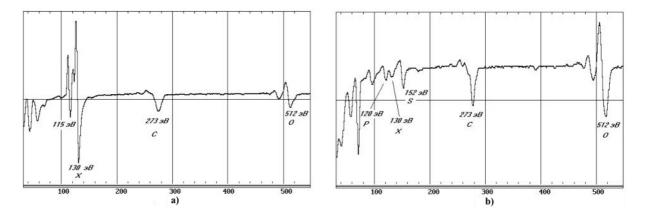


FIG. 6: Fragment of the Auger-spectrum containing the unidentifiable doublet of peaks with energies of 115 and $130 \, \text{eV}$ (a) and the fragment of the Auger-spectrum containing these peaks on the repeated registration (b).

words about how they were discovered. At first, after the Auger-spectrum registration, the peaks were erroneously interpreted as the Auger-peaks of yttrium from series MNN, because their energies well coincide. However, we were disturbed by the following circumstance: the registered peaks had high intensity, the Auger-spectrum belonged to the energy interval from 30 to 3000 eV with high exposure duration, but the brightest Auger-peak of vttrium from series LMM with an energy of $1746\,\mathrm{eV}$ was absent. In this situation, the natural question arose about whether the intensity of the 1746-eV Auger-peak of vttrium from series LMM exceeds the background fluctuations under the conditions of registration of the Augerspectrum. To solve this problem, we registered the standard Auger-spectrum of yttrium being in its simple compound Y_2O_3 under the same conditions. As a result, we established that the intensities of the Auger-peaks of yttrium with energies of 127 and 1746 eV are related approximately as 4:1. This means that if, in the situation under discussion, the low-energy doublet of peaks would belong to Y, its 1746-eV Auger-peak from series LMM must considerably exceed the background fluctuations, because the intensity of the registered doublet was very large (Fig. 6, a).

To verify that the observed doublet of peaks does not correspond to yttrium, we tried again to register it and to study its behavior and the spreading area. By returning in several days on that particle where it was observed, we failed to find the doublet. While studying the tremendous number of similar particles, we sometimes succeeded to observe the doublet in the scanning regime. However, in all the cases, its intensity was considerably less than that on the first particle, and the doublet was already absent when we made registration with a large exposure duration except for one case. The fragment of the Auger-spectrum, where it was registered for the second time, is presented in Fig. 6, b. The unidentifiable doublet is also characterized by the fact that it has no accompanying low-intensity peaks in the energy range $30 \dots 3000 \,\mathrm{eV}$. This conclusion follows from their absence in the spectrum where the unidentifiable doublet was registered with high intensity for a large exposure duration.

The fifth unidentifiable peak with an energy of 94 eV was registered on 2 specimens in 10 Auger-spectra (Table I). One of these spectra containing the peak is shown in Fig. 7, a. In all the registered spectra, its intensity

was significant and its energy position was quite exactly reproduced. Its chemical environment was the same in all cases and rather scanty: C, O, and Cu. It is typical that its environment did not contain even S, Cl, and N which are usually present in all spectra of the specimens under study. The peak was relatively stable in time and under the action of the electron beam of the probe.

We now discuss the spreading area of the unidentifiable 94-eV peak. We studied twice the specimen, where it was registered. At first, all the specimen surface and all its details were thoroughly investigated, but we did not find the peak. For the second time, the same specimen was investigated approximately in a month after secondary ion mass-spectroscopy study. On the specimen surface, we observed a significant number of craters of about 1 μ m in depth and 250 \times 250 μ m in area which appeared after the ion etching of the ion gun of a massspectrometer of secondary ions. On the bottom of craters and on the specimen surface, we observed many circular flattened light particles of $10 \dots 40 \,\mu\text{m}$ in diameter (Fig. 7, b). These particles contain always the significant amount of Pb and frequently of Si. We note that the Auger-spectra of both elements in the range $92 \dots 94 \,\mathrm{eV}$ include rather bright low-energy Auger-peaks. However, there are also high-energy Auger-peaks with energies of 2187 and 1619 eV for Pb and Si, respectively. But on some parts of the mentioned particles and only on those which were located on the crater bottom rather than on the specimen surface, we registered the Auger-spectra containing the 94-eV peak without Auger-peaks with energies of 2187 and 1619 eV. In this case, the intensity of the 94-eV peak was so large that if it would belong to Pb or Si, the Auger-spectra of both elements must contain also high-energy Auger-peaks. Since the latter were absent, we refer the 94-eV peak to unidentifiable ones. This peak is observed quite frequently, and the number of its registrations can be made as large as one likes. We note also that the 94-eV peak has no accompanying low-intensity unidentifiable peaks in the energy range $30...3000 \,\text{eV}$ (see Fig. 7, a).

The following peak can be referred to unidentifiable ones only with some reservations. It was registered only on 1 specimen and only in 1 Auger-spectrum (Table I). Its energy position can be estimated as 559...562 eV. The Auger-spectrum containing it is shown in Fig. 8. It was registered on a small light particle. Like the previous peak, it had a scanty chemical environment: C, O, and Cu. Its stability is characterized by the fact that it was not found in the repeated registration.

As for reservations, they are the following. On the one hand, the energy position of the peak can be referred to the Auger-peak of Cs from series MNN with an energy of 563 eV due to the closeness of their energies. On the other hand, it is obvious that the coincidence or the closeness of the energies of both peaks is not sufficient for such an identification. It is also necessary that the peaks of other series coincide by energies. However, the other Auger-peaks of Cs have low intensities as compared to its analytical Auger-peak (563 eV), and the spectrum under consideration was registered under such conditions that the former can to be not registered against the background (see Fig. 8). In other words, we have no sufficient reasons for to say that this peak belongs to Cs, on the one hand. But, on the other hand, this situation cannot be omitted. In any case, the peak behavior cannot be referred to the ordinary one.

By summarizing the representation of the derived experimental results, we note that one of the variants of the interpretation of the observed unidentifiable Auger-peaks which were not referred to artifacts is the assumption of their affiliation to long-lived transuranium elements, which is natural in the context of the present work. Of interest is the estimation of the atomic numbers of chemical elements, to which at least some indicated Auger-peaks can be referred: e.g., the unidentifiable Auger-peaks with energies of 172, 527, and 1096 eV.

One of the methods of estimation of the atomic numbers of chemical elements is the extrapolation based on the Moseley law [13] which gives the dependence of the energy of a definite atom level or the energy of an X-ray line from a specific atom series on the atomic number. In our situation, the Moseley law can be written

$$Z = kE^{1/2} + Z_0, (1)$$

where k and Z_0 are the constants in the limits of one series of Auger-peaks and Z is the atomic number of a chemical element whose Auger-peak from the series under consideration has the energy E.

The data on the energies of the Auger-peaks of chemical elements can be found in atlases and catalogs [10-12, 14]. It is worth to note that we are interested, first of all, in the energies of the Auger-peaks of chemical elements with large atomic numbers, but the relevant information available from the literature is rather scanty. By analyzing the literature data, we can distinguish only two clear series of bright Auger-peaks of heavy chemical elements corresponding to the Auger-transitions of series NOO. Information on them is given in Table IV, where they are conditionally denoted as Series I and Series II. By Eq. 1, we calculated the values of atomic numbers Z' with $k_{\rm I} = 2.48 \, {\rm eV}^{-1/2}$, $Z_{\rm I0} = 58.00$, and $k_{\rm II} = 4.57 \, {\rm eV}^{-1/2}$, $Z_{\rm II0} = 52.70$ for the first and second series, respectively. The deviation (see Table IV) of the rated value of the atomic number of a chemical element from its real atomic number, $\triangle Z$ testifies to that Eq. 1 describes satisfactorily the series of Auger-peaks under study. We note that a growth of $\triangle Z$ with decrease in the atomic number is related to the growing measurement error for the energy position of low-energy Augerpeaks rather than to the deterioration of the degree of the used approximation.

Now, by using the calculated constants $(k_{\rm I}, Z_{\rm I0}, k_{\rm II},$ and $Z_{\rm II0})$, we can estimate the assumed atomic numbers of chemical elements, to which the nonidentified Augerpeaks with energies of 172, 527, and 1096 eV would belong in the cases where they refer to Series I and Series

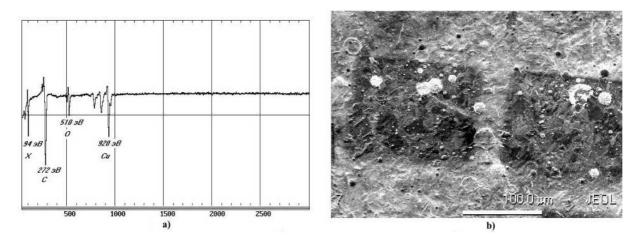


FIG. 7: Typical Auger-spectrum containing the unidentifiable 94-eV peak (a); the peak was registered on light particles in craters formed by ion etching (b).

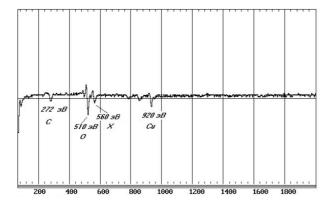


FIG. 8: Auger-spectrum containing the unidentifiable 560-eV peak (Cs).

II. The results of these calculations are given in Table V.

By analyzing the derived data, we should like to make some remarks. First of all, it is seen from Table V that the Auger-peaks with energies of 172 and 527 eV could be referred to two different series of one chemical element with atomic number in the range $112 \dots 115$. Moreover, we note as to the 172-eV peak that its affiliation to a chemical element with atomic number in the region of $90 \dots 91$ seems unlikely, because these elements have brighter Auger-peaks which were absent in the registered spectra. As to the 1096-eV peak, its affiliation to the considered series is not so realistic, probably. However, the last remark does not mean that the mentioned peak cannot be referred in a more reasonable way to some atomic number of a chemical element. This peak can be just outside of the considered series, whereas other series which could be a base for the extrapolation are not clearly manifested, unfortunately, in the available data [10-12, 14] on the energies of Auger-peaks of heavy elements.

TABLE IV: Approximation by the Moseley law of two series of the Auger-peaks of heavy chemical elements for Augertransitions of the NOO type.

Z	Element	Series I			Seri	es II	
		Energy, eV	Z'	$\triangle Z$	Energy, ${\rm eV}$	Z'	$\triangle Z$
92	U	188	92.00	0.00	74	92.01	0.01
90	Th	161	89.47	-0.53	64	89.26	-0.74
83	Bi	101	82.92	-0.08	44	83.01	0.01
82	Pb	94	82.04	0.04	40	81.60	-0.40
81	Tl	84	80.73	-0.27	36	80.12	-0.88
80	Hg	78	79.90	-0.10			
79	Au	72	79.04	0.04			
78	\mathbf{Pt}	65	77.99	-0.01			
77	Ir	54	76.22	-0.78	25	75.55	-1.45
76	Os	45	74.64	-1.36			
75	Re	34	72.46	-2.54			
74	W	24	70.15	-3.85			

TABLE V: Extrapolation of the atomic numbers of chemical elements by two series of Auger-peaks of the NOO transition.

Series	Ι	Series II		
Energy, eV	Z'	Energy, eV	Z'	
172	90.52	172	112.63	
527	114.98	527	157.71	
1096	140.07	1096	203.92	

III. CONCLUSIONS

By the results of studies of the composition of the laboratory nucleosynthesis products by local Auger electron spectroscopy, we arrive at the following conclusions.

• While studying the composition of the nucleosynthesis products on more than one hundred of specimens by AES, we registered up to several tens of chemical elements in the range from small to large atomic numbers which did not belong to the composition of the initial materials of targets and accumulating screens or, if they were present initially as impurities, exceeded the total initial content in the mentioned materials by several orders of magnitude.

- The regular appearance of the new chemical elements as reaction products, which were absent previously in the composition of the initial materials of targets and accumulating screens, is one of the facts testifying to the running of the reactions of nucleosynthesis in our laboratory setup.
- By analyzing the nucleosynthesis products, we registered 6 Auger-peaks (one doublet) with energies of 172, 527, 1096, 94, 560, and 130 (115) eV
- S. V. Adamenko, A. S. Adamenko, Proceedings of Int. Symp. New Projects and Lines of Research in Nuclear Physics, Messina, Italy, October 2002, p. 383–391 (2002).
- [2] S. V. Adamenko, Visnyk NANU (Bulletin of National Academy of Sciences of Ukraine) 2, p. 23–26 (2003).
- [3] D. Briggs, M. Seah. Practical surface analysis by Auger and x-ray photoelectron spectroscopy. Wiley, New York, 1983.
- [4] T. Carlson. Photoelectron and Auger spectroscopy. Plenum, New York, 1975.
- [5] A. Czanderna. Methods of surface analysis. v. 1 of Method and Phenomena: Their Application in Science and Technology. Elsevier, Amsterdam. 1975.
- [6] V. F. Kuleshov, Yu. A. Kukharenko, S. A. Fridrikhov, et al. Spectroscopy and Diffraction of Electrons on the Study of the Surfaces of Solids [in Russian], Nauka, Moscow, 1985.
- [7] V. T. Cherepin, M. A. Vasil'ev. Methods and Installations for Analysis of the Surfaces of Materials [in Russian], Naukova Dumka, Kyiv, 1982.
- [8] D. Woodruff, T. Delchar. Modern Techniques of Surface

which, on the one hand, are not related to the known chemical elements and, on the other hand, cannot be considered as artifacts, in our opinion. These Auger-peaks are referred by us to the class of unidentifiable ones.

• As one of the variants of interpretation of the discovered unidentifiable Auger-peaks, we consider the assumption about their affiliation to long-lived transuranium elements. By the example of the unidentifiable Auger-peaks with energies of 172, 527, and 1096 eV, we performed the extrapolational estimate of atomic numbers of the chemical elements they could belong to, by using the Moseley law.

Science. Camridge University Press, Camridge, 1986.

- [9] L. Feldman, J. Mayer. Fundamentals of Surface and Thin Film Analysis. Elsevier, Nord Holland, 1986.
- [10] P. W. Palmberg, G. E. Riach, R. E. Weber, N. C. Mac-Donald. Handbook of Auger Electron Spectroscopy. A Reference Book of Standard Data for Identification and Interpretation of Auger Electron Spectroscopy Data. Physical Electronics Industries, Inc., Minneapolis, Minnesota, USA, 1972.
- [11] T. Sekine, Y. Nagasawa, M. Kudoh, et al. Handbook of Auger Electron Spectroscopy. JEOL Ltd., Tokyo, Japan, 1982.
- [12] V. Sh. Ivanov, I. A. Brytov, V. V. Korablev, et al. Atlas of Auger Spectra of Chemical Elements and Their Compounds [in Russian], Mendeleev Institute of Chemical Technology, Moscow, 1986.
- [13] M. A. Blokhin. *Physics of X-Rays* [in Russian], GITTL, Moscow, 1957.
- [14] Y. E. Strausser, J. J. Uebbing. Varian Chart of Auger Electron Energies. Varian Vacuum Devasion. September, 1971.