Investigation of the elemental composition of samples after exposure to $\theta$-pinches in the vicinity of periodic discharges in a flow of liquid

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Abstract. The article presents the results of laser mass-spectrometric analysis of surface of the samples, exposed to long-term treatment with high current discharges in the fluid flow (water with admixtures of different composition). It has been found, that the elemental composition of the surface of the metal, in particular, oxygen-free copper, can vary considerably (up to 35 at.%) compared to the original composition (99.8 at.%) to a depth of up to 10 $\mu$m. Change of the isotopic composition (isotope concentration of 65 amu may amount to 38.1% while the normal rate is equal to 30.6%) has been indicated. Composition of water (distilled water, tap water, saline water) does not affect the results. The concentration of elements, such as Cl, may exceed the original concentration by 3,000 times. At a depth of 10 $\mu$m, the composition is similar to the original one (before the exposure).

1. Introduction
The electro-spark exposure is one of the promising methods of changing the physical properties of materials; it can be implemented by generating periodic high-voltage discharge in the fluid flow [1]. Fluid stream is used as a current-carrying electrode and a current switch, that allows generating a sequence of pulses of electrical discharge from a few nanoseconds to a microsecond with the pulse packet repetition frequency of up to several dozens of Hz.

During the primary discharge which is mostly sparking (partial discharge on the water stream surface) in the inner area of the stream occurs the pinch discharges with all signs of $z$-pinch, transforming into theta-pinches around the grounded electrode [1,2].

Found surface material modification localized in the tracks of pinch of 30 to 100 $\mu$m size inside the main discharge area about 4mm diameter. In the pinch tracks area a surface material became amorphous, indicating extreme pressure and temperature.

The method of laser mass spectrometry was used in order to examine changes in the elemental composition of the surface of various samples.

Let’s cite the results of determination of the elemental composition of the surface of the electrode, made of different metals, and then discuss the results.

2. Sample - copper plate, medium change
The conducted research regards the effect of the media, in which the discharge occurs, on the change of qualitative and quantitative composition of admixtures on the target surface. The following media have been selected for experiments: tap water, distilled water and water, specially saturated with sodium salt. The total dose of sparkling remained at the same level in the experiments, but it was impossible to keep the same interaction area. The assessments of the area for the compared experiments are: tap water - 0.5 square centimeters, distilled water - 4 square centimeters, water, saturated with salt - 0.7 square centimeters. The comparison results are presented in table 1.
Measurement was made on the EMAL - mass spectrometer with a laser-plasma ion source. The EMAL characteristic is described below:
- quantum optic generator – ILTI-203;
- crater depth, μm – 0.1 ÷ 1.0;
- crater diameter, μm – 10 ÷ 150;
- absolute lower detection limit, g – 10^{-13} ÷ 10^{-14};
- relative detection limit, % – 3\times 10^{-7};
- measurable concentration range, % – 10^{-7} ÷ 10;
- maximum detectable ion mass, amu – 500.

Table 1. Results for copper plate element analysis for different waters

| Element | Content,% wt. |
|---------|----------------|
|         | Distilled water | Tap water | Saturated salt water |
| C       | 0.4            | 0.2       | 0.2 |
| N       | 0.02           | 0.03      | 0.00 |
| O       | 0.2            | 0.7       | 0.24 |
| Na      | 0.2            | 0.2       | 0.14 |
| Mg      |                |           |     |
| Al      | 0.03           | 0.0008    |     |
| Si      | 0.07           | 0.003     |     |
| P       |                |           |     |
| S       | 0.009          |           |     |
| Cl      | 0.3            | 0.6       | 0.3 |
| K       | 0.06           | 0.07      | 0.08 |
| Ca      | 0.04           | 0.03      | 0.03 |
| Cu      | rest           | rest      | rest |

According to the comparison of the results, normalized to one area, it may be concluded that: the presence of different media does not significantly affect the quantitative composition of the elements on the surface of samples.

3. Sample - copper plate, isotope analysis
The effect of a discharge on the change of isotopic composition of the target base (copper here) has been studied. The results have been obtained on the basis of calculations of the spectra, measured with laser spectroscopy. The measurement was made by ELAN 6000 ICP-MS mass spectrometer accompanied with the sampling Laser Sampler 330. ELAN 6000 characteristic is described below:
- mass range, amu – 1 ÷ 270;
- resolution, amu – 0.3 ÷ 3;
- dynamic range – 10^8;
- relative detection lower limit, % – 10^{-6}.
As a result of the studies, it was found:
1. Isotope ratio in the copper plate without the exposure corresponds to the natural distribution of isotopes namely- portion of 65 amu is equal to 30.6%, while the tabular value is 30.9%; portion of 63 amu is the rest.
2. High isotope concentration of 65 amu: 38.1; 35.2; 36.6; 37.5 is observed in the first (surface) 2-μm layer.
3. The reduced isotope concentrations of 65 amu are observed in subsequent 2-μm layers: 2-nd layer - 28.1; 28.5; 28.8; 3-rd layer - 25.8; 26.0.
4. Three days later, high isotope concentration of 65 amu: 39.3; 38.3; 35.8; 35.3, also has been observed on a new (not analyzed previously, but exposed to the discharge) area.

4. Sample - copper plate, layer-by-layer analysis
Elemental and quantitative composition was studied both in the area of external exposure and on the unexposed surface of pure copper (99.8 at.%) sample. Areas from both front and back surfaces of exposure have been studied. In addition, a serial layer-by-layer analysis of the composition at intervals of approx. 2 µm (table 2) has been conducted.

Table 2. Results for a copper plate element analysis

| Element | Unexposed surface | Front side surface layer | Layer №2 4 µm | Back side surface layer | Layer №2 4 µm | Layer №3 6 µm |
|---------|-------------------|-------------------------|---------------|-------------------------|---------------|---------------|
| C       | 0.2               | 1.0                     | 0.28          | 2.3                     | 0.6           | 0.2           |
| N       | 0.005             | 0.1                     | 0.03          | 0.2                     | 0.07          | 0.04          |
| O       | 0.06              | 2.7                     | 0.9           | 20                      | 2.5           | 1.2           |
| Na      | 0.004             | 0.5                     | 0.3           | 0.2                     | 1.1           | 0.2           |
| Mg      | 0.003             | 0.002                   | Indefined     | 0.02                    | 0.008         | 0.004         |
| Al      | 0.002             | 0.005                   | Indefined     | 0.02                    | 0.008         | 0.002         |
| S       | 0.0007            | 0.02                    | 0.01          | 0.02                    | 0.01          | 0.016         |
| Cl      | **0.006**         | **1.0**                 | 0.6           | **11.8**                | **5.7**       | **5.4**       |
| K       | **0.005**         | **0.1**                 | **0.1**       | **0.3**                 | **0.2**       | **0.1**       |
| Ca      | **0.0003**        | **0.04**                | **0.03**      | **0.04**                | **0.02**      | **0.03**      |
| Cu      | rest              | rest                    | rest          | rest                    | rest          | rest          |

As a result of the analyses of the composition, the following features of the studied samples can be pointed out:

1. No formation of new elements, absent in the original sample, is observed.
2. On the surface of samples, in places subjected to external exposure, an increased concentration of admixtures, namely C, N, O, Na, Cl, K, Ca is fixed.
3. The more concentrated exposure is, the greater the concentration of the specified admixtures is. The concentration of exposure determine as estimated energy density to the unit of the irradiated plate surface. Averaging over surface area is determined by density of the craters corresponding size (30 to 100 µm diameter) which is function of discharge intensity, repetition frequency and exposure time.
4. The concentration of certain admixtures, such as Cl, exceeded the content compared to the initial one by 3000 times, and the concentration of Na exceeded the initial concentration by 300 times.
5. The concentration of specified admixtures decreases with deepening in the sample surface.
6. In terms of the majority of the admixtures, concentration of elemental composition of the sample returns to the original one approx 10 microns below the surface layer.
7. Concentration of admixtures in the atomic percentage on the surface, subjected to external exposure, exceeds one third (up to 35 at. %) as compared to the originally pure copper (99.8 at.%).

5. Sample - copper plate in a 3 weeks’ interval

Elemental and quantitative composition was studied both in the area of external exposure and on the unexposed surface of the same copper sample. Areas from both front surface and back surfaces of exposure, where the traces of exposure had been observed, have been studied. 3 weeks later the surface analysis was redone. This time new areas, located near the previously analyzed ones, were analyzed (table 3).

Table 3. Results for copper plate in a 3 weeks’ interval
As a result of the composition analyses, the following features of the studied samples can be pointed out:

1. 3 weeks after the external exposure, new elements, not contained in the original sample, such as fluoride and phosphorus, emerged on the sample, subjected to external exposure.

2. On the samples surfaces, in places subjected to the external exposure, the increased concentration of admixtures, namely C, N, O, Cl, has been generally decreased. The number of elements, such as Na, K, Ca, either does not change or increases.

3. As compared to the surface, free from the exposure, 3 weeks after the exposure, there is still a high content of previously specified elements on the surface.

6. Sample - beryllium plate
Elemental and quantitative composition was studied both in the area of external exposure and on the unexposed surface. In addition, a serial layer-by-layer analysis of the composition at intervals of approx. 10 µm has been conducted. Studies of the composition have been resumed 24 hours later at the same place, deeper in the surface (table 4). Layer №4 correspond to original material composition of the plate.

Table 4. Result for a beryllium plate

| Element | Unexposed surface | Front side surface layer | Front side. Surface layer. After 3 weeks | Back side surface layer | Back side surface layer. After 3 weeks |
|---------|-------------------|--------------------------|------------------------------------------|------------------------|----------------------------------------|
| C       | 0.2               | 1.0                      | 0.2                                      | 2.3                    | 0.8                                    |
| N       | 0.005             | 0.1                      | 0.04                                     | 0.2                    | 0.03                                   |
| O       | 0.06              | 2.7                      | 1.5                                      | 20                     | 3.2                                    |
| Na      | 0.004             | 0.5                      | 0.5                                      | 0.2                    | 0.8                                    |
| F       |                   |                          | 0.001                                    |                        | 0.008                                  |
| Al      | 0.003             | 0.002                    | 0.004                                    | 0.02                   | 0.004                                  |
| Si      | 0.002             | 0.005                    | 0.005                                    | 0.02                   | 0.012                                  |
| P       |                   |                          | 0.01                                    |                        | 0.002                                  |
| S       | 0.0007            | 0.02                     | 0.01                                    | 0.02                   | 0.006                                  |
| Cl      | 0.006             | 1.0                      | 0.6                                      | 11.8                   | 0.4                                    |
| K       | 0.005             | 0.1                      | 0.09                                    | 0.3                    | 0.2                                    |
| Ca      | 0.0003            | 0.04                     | 0.02                                    | 0.04                   | 0.2                                    |

As a result of the analyses of the composition, the following features of the studied beryllium samples can be pointed out:

1. No formation of new elements, not contained in the original sample, is observed.
2. The increased concentration of oxygen only was found on the sample surface, subjected to the 
   external exposure, which was reduced to the initial level (free of exposure, i.e. the clean surface) 
   deeper in the surface.
3. The concentration of boron on the surface, subjected to exposure, decreases (by about 10 
   times).
4. Comparing the balance of content of oxygen and boron (decrease of one element and increase 
   of another element in atomic percents), it can be said, within the limits of the error, that their total 
   number before and after the exposure remains unchanged.
5. It should also be noted, that there are no effects of increased content of admixtures, which 
   were observed after the exposure to the copper plate.

7. Sample - tungsten plate
Elemental and quantitative composition was studied both in the area of external exposure and on the 
unexposed surface.

Table 5. Result for a tungsten plate.

| Element | Content, % wt. |
|---------|----------------|
|         | Unexposed surface | After exposure, the first layer | After exposure, the second layer | After exposure, the third layer | After exposure, the fourth layer |
| H       | 0.3              | 0.5                        | -                           | -                            | -                          |
| C       | 0.6              | 0.2                        | 0.1                         | 0.08                         | 0.08                       |
| N       | 0.008            | 0.02                       | 0.008                       | 0.004                        | 0.001                      |
| O       | 2.6              | 1.5                        | 0.2                         | 0.08                         | 0.07                       |
| Mg      | 0.08             | 0.35                       | 0.12                        | -                            | -                          |
| Na      | 3.0              | 0.5                        | 0.3                         | 0.04                         | 0.009                      |
| F       | 0.04             | 0.02                       | -                           | -                            | -                          |
| Al      | 0.1              | 0.16                       | 0.08                        | 0.012                        | 0.009                      |
| Si      | 0.3              | 0.20                       | 0.08                        | 0.015                        | 0.009                      |
| P       | 0.01             | 0.020                      | -                           | -                            | -                          |
| S       | 0.1              | 0.1                        | 0.02                        | -                            | -                          |
| Cl      | 0.2              | 0.6                        | 0.3                         | 0.05                         | -                          |
| K       | 2.0              | 0.25                       | 0.14                        | 0.025                        | -                          |
| Ca      | 0.4              | 0.25                       | 0.13                        | 0.020                        | -                          |
| Ti      | 0.030            | 0.015                      | -                           | -                            | -                          |
| Cr      | 0.004            | 0.004                      | -                           | -                            | -                          |
| Mn      | 0.007            | 0.006                      | -                           | -                            | -                          |
| Fe      | 0.06             | 0.15                       | 0.09                        | -                            | -                          |
| Ni      | 0.015            | 0.003                      | -                           | -                            | -                          |
| Cu      | 0.01             | 0.3                        | 0.28                        | -                            | -                          |
| Zn      | **0.01**         | **0.2**                    | **0.1**                     | -                            | -                          |
| Mo      | 0.12             | -                          | -                           | -                            | -                          |
| W       | rest             | rest                       | rest                        | rest                         | rest                       |

As a result of the analyses of the composition, the following features of the studied samples 
can be pointed out:
1. On the surface of samples, in places subjected to external exposure, a decrease of 
   concentration of admixtures, namely C, N, O, Na and K was observed.
2. The concentration of Cu and Zn, increases significantly (30 times and 20 times 
correspondingly).

8. Discussion
The analysis of the obtained results and the results of the materials research and theoretical works, 
devoted to the study of nature of pinch plasmoids (observed at used in this work periodic high-voltage
discharge in water), allows to make a preliminary assumption about possible monopole magnetic charges formation [3,6,7] during such discharges. This monopole charges have been searched earlier in the assumption of their separation by strong magnetic fields. At a pinch base of 30 μm in diameter and currents up to several kA recorded in experiments, the formation of a vertical magnetic field with the induction of several dozen T, separating and accelerating the monopoles, is not excluded [4,5]. The changes of element composition in the surface layer from the back (!) side of the sample (few mm thick) can be assumed to be an indirect confirmation of this hypothesis. The possible explanation of this fact may be the distribution of similar charged monopoles (“northern” monopoles only or “southern” monopoles only), subjected to mutual repulsion. The monopole overrunning the sample would require additional measures (extremely strong magnetic field) [8]. Within the area of monopole localization, the elemental composition should normally be changed presumably due to extra-high density of electrons nearby (which can exceed the electron density in the environment by several orders of magnitude). At such extra high density of electrons the neighboring nuclei fuse together, forming the heavier ones, since the Coulomb barrier for nucleus is decreased under influence of negative charge of these electrons (example - formation of phosphorus and fluorine out of air atoms and ions of hydrogen and oxygen[5,9]). At this conditions neutronization, i.e. fusion of electron and proton in a nucleus, should occur even more easier. Atom can relax over time to its original state with electrons leaving the surface in the process of beta decay [9]. Glowing of the surface in separate spots corresponding to beta decay was reported earlier, or remains in the form of a new element (neutralization of the nearest isotope nucleus), which, presumably, can be facilitated by the probing laser radiation.

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