Self-propagating high-temperature synthesis of the high-current emission lanthanum and niobium contained ceramics

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Abstract. The paper describes the production of metal-ceramic high-current emitters containing lanthanum hexaboride LaB$_6$ by self-propagating high-temperature synthesis. Tests of emitters as explosive-emission cathodes in an OMEGA-350 microsecond accelerator are presented. Experiments show that when a metal-ceramic cathode is used, the emitted energy was 12-17% higher than that produced by tungsten and graphite cathodes and the beam “signature” is a circle with a small (~ 15%) variation in radius.

1. Introduction
Accelerators of particle occupy a very important place in the industry. The most important element of devices generating flows of charged particles is cathode [1]. Cathodes made from lanthanum hexaboride (LaB$_6$) are widely used in accelerators to generate high-current electron beams in stationary and quasi-stationary modes. However, sintered pellets of LaB$_6$ are brittle and easily destroyed by heat shock [2,3]. Consequently cathodes with layers of LaB$_6$ plasma-sputtered onto substrates made from high melting-point metals are commonly used.

Here we present results of the synthesis of high-current emitters for explosion-emission cathodes containing LaB$_6$. The cathodes are produced by self-propagating high-temperature synthesis [4].

2. Experiment
The initial reagents are powders of La and Ti oxides, amorphous B and Si. The blend is additionally ballasted with a reactive stoichiometric mixture of Ni and Al powders taken in various proportions, thus the emitters are tougher due to the formation of a Ni-Al matrix containing inclusions of LaB$_6$ and TiB$_2$ compounds. The mixture is ground in a ball mill and they dried in a low vacuum. Emitter pellets are produced by moulding the mixture in press-molds of various diameters (up to 30 mm).

The explosive reaction is initiated at the edges of the pellet by heating them in a quartz tube furnace in air. Temperature diagrams generated by thermocouples placed on the sides and upper edges show that the process begins as surface combustion. After the combustion front has passed over the surface, a secondary heat explosion reaction begins inside the sample. The surface combustion acts as a “chemical furnace” that heats the center of the pellet.

The explosion consists of two reactions, viz.,

$$\text{TiO}_2 + \frac{16}{3} \text{B} \rightarrow \text{TiB}_2 + \frac{2}{3} \text{B}_2\text{O}_3,$$

$$\frac{1}{2} \text{La}_2\text{O}_3 + 7\text{B} \rightarrow \text{LaB}_6 + \frac{1}{2} \text{B}_2\text{O}_3,$$

with adiabatic combustion temperatures of 1400 and 1300 K, respectively [4], together with the synthesis of nickel aluminides.
In order to test the emitters, several experiments were performed using the OMEGA-350 microsecond high-current electron beam accelerator. The accelerator OMEGA-350 allows generated electron beams with duration from 1 to 3 microseconds at a current density of 50 kA/cm$^2$ and energy of 350 keV electrons to ensuring full supply of energy of the beam more than 1 kJ generation efficiency to 80%. The complex includes a high-current vacuum diode, voltage pulse generator, and the system of collecting and processing information. Voltage pulse generator is a seven cascade generator, assembled in a classic case Arkadiyeva-Marksa. With voltage pulse generator may exercise the power vacuum diode or other physical equipment pulse voltage with a maximum peak value of 350 kV, negative or positive polarity in the manual (single) or automatic (multiple) modes.

3. Results and discussion
The temperature developed on the pellet surface during the surface phase is 1350-1450 K and then rises to 2200 K when the secondary heat explosion reaction starts. The synthesis is initiated when the surface temperature is over 1000 K.

The structure of the emitter surface is shown in figure 1. Several compositions were used to produce the emitter, viz. $\text{La_2O_3}$ (D 543 NTD OST 4S-194-81); $\text{TiO_2}$ (TU 6-09-2166-72); B (TU 113-12-132-83); Si (KR). The mixture is ballasted with powders of nickel (PNK-OT1) and aluminum (PA-4).

![Figure 1. Electron-microscope photograph of the high-current emitter’s surface (magnification 1:1000, E-modulation).](image-url)
Many inhomogeneities (LaB$_6$ and TiB$_2$) can be seen on the emitter surface; some of the Si reacts with the TiO$_2$ to form the silicides TiSi, TiSi$_2$, and some Si forms inclusions 10-25 µm in size. These are uniformly distributed over the volume and surface.

Lanthanum hexaboride LaB$_6$ has a considerable field-emission current. The work function is considerably lower in the presence of La atoms in the surface layer of the emitter [5]. The LaB$_6$-Me(IV)B$_2$ compounds (where Me(IV) indicates a group IV metal, i.e., Ti, Zr, Hf) have relatively high thermal-emission characteristics.

The initial stages of the explosion emission involve thermal and field emissions. Field- and thermal-emission currents lead to the Joule heating of small points and to a subsequent explosion. Given the good field- and thermal-emission characteristics of materials, the time between the voltage pulse and beginning of the explosion can be shortened and, as a consequence, the time needed for the cathode plasma to form shortened. This, in turn, shortens the delay between the electron-current signal and the accelerating voltage pulse, and as a result, the energy transferred from the diode to the beam is higher.

The Si inclusions on the emitting surface increase the electric field at their boundaries. Thus, when there is no emission from some points, adjacent points stimulate explosive emission due to the potential differences that arise due to the currents passing through the points. As a result, the time needed for the plasma flares to merge on the emitting surface into a uniform plasma layer is shorter and the emitted electron beam is more uniform over its cross section.

Next step of experiments is test to emitters on the OMEGA-350 microsecond high-current electron beam accelerator.

The following figure shows oscillograms of accelerating voltage and beam current pulses outputted to the surface of the irradiated target.

Figure 2 and figure 3 show the results for the regime with current pulse duration of 2.4 microseconds.

![Figure 2. Accelerating voltage U and beam current I pulses.](image-url)
Figure 3. Beam power $W$ within current pulse duration.

We obtained the following beam parameters:

- Beam energy storage $814$ J.
- Beam generation efficiency $71\%$.
- Energy density on irradiated surface $65\ldots70$ J/cm$^2$.

Figure 4 and figure 5 show the results for the regime with current pulse duration of $1.75$ microseconds.

Figure 4. Accelerating voltage $U$ and beam current $I$ pulses.
We obtained the following beam parameters:

- Beam energy storage 808 J.
- Beam generation efficiency 68%.
- Energy density on irradiated surface 65…70 J/cm².

Also in experiments we used detectors of absorbed energy. The detector was a solid-state chemical dosimeter, viz., polycrystalline potassium nitrate in the form of pellets 16 mm in diameter and wide enough to absorb the beam fully. The density of the energy absorbed by the dosimeter in one pulse was calculated from the radiation-chemistry yield of nitrate ions \([NO_2^-]\). Experiments show that when a metal-ceramic cathode is used, the emitted energy was 12-17% higher than that produced by tungsten and graphite cathodes.

4. Conclusions
The homogeneity of the cathode plasma was assessed from the “signature” of the electron beam output from the diode. The ES spectrographic photoplates and alkali-halide crystals were used as the sensitive elements. Experiments show that when a metal-ceramic cathode is used, the beam “signature” is a circle with a small (~15%) variation in radius. The saturation of the crystal color is homogeneous within the circle. The negative picture of the “signature” is also homogeneous. The reproducibility is significantly worse when a tungsten cathode is used. The shape of the “signature” stochastically deviates from the circular to elliptical, and in some cases the beam fragments are observed. The explicit color inhomogeneity inside the “signature” is evidence of fragmentation.

References
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