Investigation of nanostructural and monocrystalline materials for units of high intensive X-ray sources

A A Pavlov¹, M L Taubin¹,², D A Chesnokov¹ and A A Yaskolko¹,²

¹Federal State Unitary Enterprise "Science Research Institute "LUCH", Zheleznodorozhnaya st, Podolsk, Moscow region, 142100, Russia
²National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe highway 31, Moscow, 115409, Russia

E-mail: taubin@sialuch.ru

Abstract The concept of increase of characteristics x-ray tubes by use nano-and monocrystal materials is offered in the report.

1. Introduction
Results of own works of the authors and works of the Russian, Japanese, Korean and American experts in refractory metals nanocomposite with increased strength characteristics for cathodes and anodes are analyzed in this report. It is shown, that though monocrystals have shown the better characteristics in comparison with traditional polycrystals both as anodes and as cathodes, more radical way of increase of operational characteristics of the x-ray tubes is application refractory nanostructural materials.

The results of experiments showed that high-temperature sample annealing resulted in a sharp increase (by a factor of more than 3) in material creep resistance at a temperature of 2200°C.

The study of emission characteristics shows that single-crystal tungsten with crystallographic orientation of emitting plane (111) has higher emission characteristics (work function equals 4.43 eV) than polycrystalline tungsten. But best results has nanoscale W.

In this work results of research of evaporation of tungsten are presented. It is shown that the monocrystal has essential advantages on this parameter. It is important for materials working at temperatures more 2000°C.

An increase in the emission characteristics of a cathode and the strength characteristics of an anode at the same time can be achieved by modifying standard tungsten powders (for cathodes), tungsten and molybdenum powders (for anodes) by nanoscale powders of refractory materials (grain size is 10-100 nm). W, Mo, Nb, Re nanopowders are proposed to be used for producing anodes based on tungsten and molybdenum. It must increase allowable unit loads of an X-ray tube by reducing porosity, increasing strength and plasticity of anode material. Prerequisites for these proposals are some results of work where an increase in density, strength by 10-30%, and plasticity by a factor of 1.5 - 2 was achieved due to modification of steels by ultra-disperse powders of refractory compounds.

The authors carried out some experiments to determine the creep of tungsten alloyed by...
potassium on samples as foils 100 μm thick, used when manufacturing X-ray tube cathodes. The results of these experiments showed that high-temperature sample annealing resulted in a sharp increase (by a factor of more than 3) in material creep resistance at a temperature of 2200°C. The authors think that this effect results from forming nanoscale little bubbles filled with potassium at the grain boundaries. Results of this work can be used when developing X-ray tubes with the increased operational characteristics.

2. Emission properties of tungsten

The problem of X-ray tubes at the present stage of developing medical equipment consists in the necessity of producing X-radiation of high intensity when providing small-size focal spots.

It is of particular importance for X-ray tubes of digital scanning X-ray units and mammography. Cathode problems are key problems for providing required roentgen optical characteristics of X-ray tubes.

The main task for having a focal spot of necessary size is electron focusing. Today, the most promising way of producing small-size focal spots is applying the design of cathodes with flat emitting surface.

A flat-shaped emitter is more efficient as compared with a spiral as it has more uniform electric field distribution and, also, large area which is important for producing X-radiation of high intensity [1]. A decrease in work function for increasing emission current is only possible when using new materials.

The results of many-year investigations of materials conformably to their application as emitters of thermionic energy converters [2, 3] showed that tungsten with single-crystal structure could also be promising material for X-ray tube emitters. The results of these investigations showed higher emission characteristics, lower and uniform (as compared with polycrystalline) tungsten evaporation from surface and higher dimensional stability at high operating temperatures due to absence of recrystallization (a considerable disadvantage of emitters made of polycrystalline) up to melting point as well. Proceeding from the above-mentioned, we think that producing a cathode with a flat emitter made of single-crystal tungsten is one of the promising trends in the area under consideration.

The use of single-crystal tungsten as emitter material allows, firstly, reducing the emitter working temperature by 70°C as the work function of the crystallographic face [111] of single-crystal tungsten is (4.4±0.05) eV, being lower by 0.15 eV than the work function of polycrystalline tungsten (4.55±0.05) eV, secondly, making easier electron focusing due to lower emission contrast range of a single crystal.

The results of studying the emission characteristics (Figure 1) show that single-crystal tungsten with crystallographic orientation of emitting plane (111) has higher emission characteristics (work function equals 4.43 eV) than polycrystalline tungsten. However, the best results have nanoscale W (Figure 1).
3. Compare of damageability of poly- and monocrystalline tungsten

The second task for increasing the output characteristics of X-ray tubes is anode problems, i.e. a decrease of its failure.

Doping of tungsten by rhenium is not always optimum, as to mammography it is not acceptable. In this connection, the authors have offered manufactured and tested single-crystal molybdenum (for mammography) and single-crystal molybdenum coated with single-crystal tungsten (for other applications) as anode material.

It is shown that the dependence of reducing in X-ray tube dose rate for a single-crystal anode (Figure 2) differs slightly from the typical dependence for a polycrystalline molybdenum anode. After reducing the dose power by 10% during the first 1500 switches, stabilization takes place, providing large service-life of an X-ray tube.

In work results of research of evaporation of tungsten also are presented. It is shown that the monocystal has essential advantages on this parameter (Figure 3). It is important for materials working at temperatures more 2000°C.
4. Advantage of nanomaterial for using in X-ray units

Though single crystals showed better characteristics both as anodes and as cathodes, the application of nanomaterials may be a more radical way of increasing the operating characteristics of X-ray tubes.

For instance, the application of nanopowders (Mo, Nb) for producing anodes based on molybdenum or molybdenum alloy with niobium must exclude the influence of the technological disadvantages above-described and increase permissible unit loads to the focal spot due to a decrease in porosity, an increase in strength and plasticity of material.

Some investigation results are known [4] where the work function of nanocrystal tungsten was lower by 0.8 eV than that of coarse tungsten. This phenomenon can be explained by the fact that when forming nanomaterial, current tubes with lower work function appear in the range 10 nm wide near grain boundaries. This effect can considerably increase X-radiation intensity at the same cathode temperatures or decrease the cathode temperature at the same X-radiation intensity by 400°C. Estimations show that at the temperature at which the effect of an increase in the emission characteristics for this material is still available (T ≤1500°C), one may rely on obtaining the density of emission current equal to 10 mA/cm².

A number of publications have appeared lately where some results of developing X-ray tubes with the use of carbon nanotubes (CNT) as cathode materials are described.

The development engineers from the University of South Caroline together with the other researchers presented materials on developing a scanning source of X-radiation with a cathode made of CNT. The anode voltage was 40-60 kW, electric field strength - 2.3-3.1 V/cm, emission current density reached 1 A/cm².

In parallel with the improvement of the characteristics of cathode made of CNT, the application of nanodimensional dopes may also increase strength properties of tungsten used conventionally.

5. High temperature deformation of tungsten

We obtained the effect of an increase in tungsten strength characteristics at high temperature annealings of samples as 100 μm thick foils of BA type tungsten (tungsten with silicon-aluminum and alkali additives) (Figure 4).
Investigations in the structure of this material samples after annealings (Figure 5). Allowed finding out nanodimensional ($\leq 200 \text{ nm}$) inclusions along the grain boundaries of tungsten subjected preliminarily to heat treatment, resulting in a sharp increase (by a factor of more than 3) in material creep resistance at a temperature of 2200°C.

**Figure 4.** Deformation dependence on time at creep tests. 1 - polycrystalline tungsten without heat treatment; 2 - polycrystalline tungsten preannealed at a temperature of 2500°C for 5 hours. 3 - polycrystalline tungsten preannealed at a temperature of 2500°C for 7 hours. 4 - monocryalline tungsten.

**Figure 5.** Microstructure of polycrystal tungsten subjected to heat treatment at a temperature of 2500°C for 5 hours.

As a whole, a qualitative increase in the operating characteristics of X-ray tubes with the use of nanomaterials and monocrystal materials can be achieved by a simultaneous increase in the density of electron current (a flat cathode of nanotungsten or tungsten nanocomposite) and by an increase in load-carrying capacity of an X-ray tube (anode made of nanocomposites of molybdenum with niobium or tungsten with rhenium) by providing a great number of interactions between electrons of increased density and a great number of atoms of anode target materials.

For an establishment of influence of duration of annealing on thermal stability of structure of a material on samples, of annealing within 30 minutes, the diamond pyramid puts marks. Then, these samples were annealing at temperature 2400°C within 1.5 hours and 3 hours.
Thus, temperature stability of this effect as shown is at high (up to 2400°C) temperatures. The similar effect of formation nanosizes pores in wires of modified tungsten is described in a number of works [4-6].

6. Conclusions
Comparison of structure and properties of samples from foils with equal and nonequal structure, past preliminary heat treatment and without it is carried out. Thus, for the first time it is received nanocompozite of tungsten in the form of a foil. Structure of this material is stable up to temperatures 2200 °C.

The effect of an increase in tungsten strength characteristics at high temperature annealing of samples as 100 μm thick foils of BA type tungsten (tungsten with silicon-aluminum and alkali additives) obtained.

From experimental data it is visible, that speed of creep of a sample past preliminary thermal processing is lower, than for sample without preliminary thermal annealing. The similar behaviour of a material is connected to distinctions in structure of the tested samples.

The results presented are a cause for developing a new generation of X-radiation sources with increased diagnostic opportunities.

Acknowledgements
This work was performed within the framework of the Center of Nuclear Systems and Materials supported by MEPhI Academic Excellence Project (contract № 02.a03.21.0005, 27.08.2013).

References
[1] Alekseev S, Taubin M and Yaskolko A. 2009 20th Workshop ISTC in Korea Nanomaterials and nanotechnology (Seul)
[2] Alekseev S V, Taubin M L and Yaskolko A A 2008 Proc. 2 All-Russia national congresses on radiology
[3] Alekseev S V, Taubin M L and Yaskolko A A 2008 Medical physics 2
[4] Wright P K. 1978 Metallurgical Transactions A 9A 955
[5] Yamazaki S, Ogura S, Fukazawa Y and Hatae N. 1978 High Temperature - High Pressure 10 329
[6] Briant C L Potassium 1993 Metallurgical Transactions A 24A. 1073
[7] Klinkov A E, Nikolaev Yu V and Vybyvanetz V I 1991 Proc. The second Inter.Confer. on Nuclear Power Engineering in Space. Physics of Thermionic Energy Converters (Sukhumi) 37