DEVELOPMENT OF NSC KIPT NUCLEAR MICROPROBE

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Development of the microanalysis methods is stimulated by investigations of actual materials. Their properties depend not only on an atomic structure, but also on imperfections of this structure, phase composition, areas of impurity accumulation, various kinds of microinclusions etc. Characteristic sizes of these objects are usually ranged from a few parts of micron to several tens of micron. So, there is need for methods which provide the analysis of chemical elements in samples with the corresponding level of spatial resolution.

In the development of semiconductor materials and materials for microelectronics, in laser engineering, in the production of various kinds of composite materials and alloys with multiphase structure, and also in investigations of geological objects and biological systems on microlevel, the progress would be impossible without use of microanalysis methods.

A part of these methods is used mainly in the surface analysis (X-ray spectroscopy, Auger-electron spectroscopy, spectroscopy of scattered slow ions, mass-spectrometry of secondary ions), because the thickness of an analyzable layer in these methods is insignificant (0.0005-0.01 microns). An application of these methods to volume analysis using layers sputtering leads to an essential deterioration of depth resolution because of sputtering non-uniformity.

It is necessary to notice that the sample-destructive methods are often undesirable, since after their application the sample becomes unsuitable for other researches, and a preparation of the samples is a laborious procedure. Besides, some samples are unique (such as samples of cosmic origin substance, objects of archaeology etc.).

For studies of chemical elements distribution over a volume are more widely used the microanalysis methods which provide the greater depth of the analysis. The laser mass-spectroscopy is used frequently among destructive methods of analysis. The most popular nondestructive methods are the electron probe microanalysis (EPMA) and analysis by means of the nuclear microprobe (NM).

In EPMA an excitation of X-rays is produced by an electron beam of energy 10…100 keV. As the electron beam disperses during penetration into sample, the lateral resolution at the surface ranges usually from 0.5 to 5 microns for the given method, in spite of the fact that an initial beam diameter can be by 2-3 order of magnitude less [1]. The limit of mass concentration determination is about 0.5% for the X-ray spectrometers with energy dispersion and ~ 0.1% for ones with wave dispersion [1]. The sensitivity of the method is limited by a high level of bremsstrahlung accompanying the electron energy dissipation. This method is widely used despite of its low trace element detection limits because it is embodied in rather compact commercially available devices.

The first NM was constructed by Cookson in 1970 [2]. In this microprobe 3 MeV proton beam was focussed to a diameter of a few microns by means of quadruplet of quadrupole lenses ("Russian quadruplet").

The NM has the following advantages in comparison to the electron microprobe: 1) the spatial resolution is practically determined only by diameter of the focussed beam on a sample because the beam of protons or other accelerated ions with energies of a few MeV scatters weakly in a sample; 2) the NM allows to carry out investigations in parallel by several nuclear analysis methods (NAM) which is well-known as quantitative methods for a long time [4,5]; 3) the limit of impurity determination by means of induced X-rays registration (PIXE-method) is by several order less than that for the electron microprobe, because the NM has the low bremsstrahlung background; 4) it is capable to analyze elements with A=1-14 by means of nuclear reaction (PIGE, NRA), while it is a rather complicated problem for the standard electron microprobe; 5) it is possible to carry out the analysis of layers and structures along depth of a sample by means of Rutherford backscattering (RBS).

Unfortunately, the NM did not become a serial device and it will hardly become that in the near future. The reason is that NM is bound to an accelerator (as a rule, to an electrostatic one, because this type of accelerators has the best energy stability). The accelerator-microprobe complex is a rather expensive facility and it requires large areas for its arrangement (sometimes a separate building.).

So, there is the tendency to develop the NM using available accelerators and existing experimental areas. As a rule, such complexes are developed in the research centers and large university labs.

In 1995 there were about 30 high energy NM with the beam energies exceeding 1 MeV in the world [6]. At some facilities a submicron spatial resolution for a high microbeam current (100 pA) was achieved [6-8]. The scanning transmission ion microscope (STIM), based on the NM [9, 10], was developed and a resolution of 0.05 μm was achieved [11]. In STIM the beam currents are low and each ion passed through a sample is recorded.

Thus, the NM practically has no competitors among other non-destructive techniques, both in the detection sensitivity and in the range of analyzable elements, for performing microanalysis with a resolution from submicron values to a few tens of micron.

In NSC KIPT the activities on the NM development were started in the middle of eighties. The calculations of the beam formation system, the parameters of NM and examples of investigations performed with the NM are discussed below.

The scheme of ion beam formation system is given in the Fig.1. The accelerated ion beam, collimated by a pair of diaphragms, passes through the focussing system and hits a sample. The focussing system based on magnetic quadrupole lens doublet was chosen. The doublet of lenses is the simplest system of strong

focusing, that is important for adjusting and running the system in a routine operation. Besides, a doublet of lenses ensures the highest ion currents, as compared to a triplet and a quadruplet, at the given length of system, the lens geometry and the size of the focussed beam on the target [12].

The disadvantage of doublet consist in different values of the demagnification coefficients M_x^{-1} and M_y^{-1} in two orthogonal planes. However, it can be compensated by the proper choice of the sizes of rectangular diaphragms (object and angular).

Let's dwell on some aspects of the executed calculation and assume that Z is the longitudinal axis of the lens, XZ and YZ are, accordingly, the focusing and defocusing planes of the quadrupole lens, q and p are, accordingly, the charge and the momentum of a beam particle. Then the particle trajectory in the field of the lens is described by the following system of two nonlinear differential equations for coordinates of a particle in the transverse plane, $x(z)$ and $y(z)$ (see, for example, [12]):

$$\begin{cases} x'' + \dfrac{q}{pc}\sqrt{1 + x'^2 + y'^2}\,[(1 + x'^2)B_y + x'y'B_x - y'B_z] = 0 \\ y'' - \dfrac{q}{pc}\sqrt{1 + x'^2 + y'^2}\,[(1 + y'^2)B_x + x'y'B_y - x'B_z] = 0 \end{cases}$$

where c is the speed of light, B_x, B_y, B_z are the components of magnetic field of lenses..

The following approaches to the trajectory calculation are possible: 1) calculation in linear approximation, when all nonlinear terms in these equations and in the expression for a lens field are omitted; 2) calculation using a 3-rd order expansion of these equations by entrance coordinates x_0, y_0 and slope angles x_0', y_0' (the values of coefficients are obtained in works [13, 14]); 3) numerical integration of the system of nonlinear equations.

To ensure the maximum precision in calculations we used the last approach. The terms up to 6-th order in the field expansion were taken into account. They appear as the result of a substitution of an ideal hyperbolic shape of the "working" surface of poles with more technological – cylindrical [15] and a breakage of the "working" surface [16].

The decreasing of the field gradient $G(z)$ near the face ends (fringing field) was described by a bell-shaped function [17]. Three components of the fringing fields were defined using Taylor expansion in terms of x and y, where expansion coefficients are expressed through $G(z)$ and its derivatives.

Chromatic factor was determined by varying input particle energy.

Thus, the developed codes took into account practically all significant types of aberrations: a) chromatic; b) aberrations due to the finite angular size of the beam (the special case is the spherical aberration); c) fringe aberrations, caused by the axial field components near the face ends of lenses; d) aberrations, arising from the non-linearity of lens field. These codes allow to compute both the total effect of all aberrations, and contributions of separate aberrations to

the beam size and the shape of the focussed beam on a target.

The main goal of calculation of the beam forming system is to ensure the maximal phase space and, as a result, the maximal beam intensity for the given size of the beam on a target. Since some parameters of the system can not be optimized proceeding only from the requirement of phase volume maximality. So, for example, it is desirable distance a (see, Fig. 1) be more longer, but it limits by size of workroom, where installation is supposed to dispose. On other hand, distance b should be minimum, but it is necessary to leave space before the target for arrangement of detectors. Energy E of particles and its relative instability dE/E also are objective parameters of the accelerator.
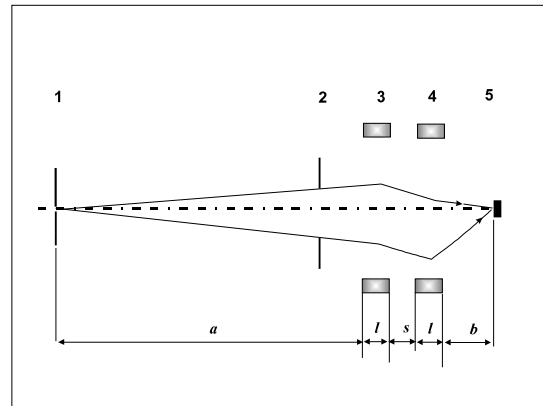

Fig.1.The NM ion beam formation system on base of magnetic lenses doublet. 1-object diaphragm, 2 - angular diaphragm, 3-4 - lenses, 5 - target, a - distance between object diaphragm and 1-st lens, l - effective length of lens, s-distance between lenses, b - distance between 2-nd lens and target. The envelope of beam in horizontal and vertical planes is showed.

If these parameters and the parameters of lenses are set then it is possible to achieve a maximum phase space by varying the sizes of collimator diaphragms. The program of optimization of the diaphragm parameters (4 parameters) with 2 constraints imposed on these parameters (the fixed sizes of the beam on a target in x- and y- directions).

Taking into account the layout of ESU-4,5 accelerator, its energy stability and the space required for placing detectors in front of the target, the following parameters for calculation were selected: a =2500 mm, b=100 mm, dE/E = 0,05 %. The calculation was executed for a "classic" version of doublet ($s=l$) and proton energy of 2 MeV, (the energy can be increased up to 3,5 MeV). The calculations were executed for a wide range of values of lens effective length.

As the result of these calculations the optimal value of 50 mm for the lens effective length was obtained. For this effective length and the value of lens aperture radius of 6,5 mm it is possible to focus the beam if the field induction at the pole does not exceed 0,4 T (such value is far from saturation and is easy provided).

Following step was determination of installation tolerances of poles in lenses at their assembly. For this purpose the aberrations of system, arising from the

poles displacement were investigated [18]. It was shown that the errors in pole installation of 5 μm lead to decreasing the sizes of collimator diaphragms , and, consequently, to a loss of beam intensity in 2 - 3 times if the beam spot size at the target is kept within 1×1 μ m^2.

Indispensable alignment element of a doublet is rotation of lenses and their displacement in the plane transversal to the beam axis. The effect of these manipulations on field distribution in the focusing system was studied in ref. [19]. The expression for disturbed field distribution from this work we used for calculation of distortion of focused beam spot on target. It was shown that the rotation misalignment of lenses along the longitudinal axis 0.05 mrad decreases the beam intensity in 2-3 times. The requirements for the lens rotation around of transverse axes and their displacement in the transverse plane are not so critical.

The detailed description of the calculation of ion optical system and the optimization of its parameters can be found elsewhere [20-22].

Let's stay briefly on the parameters of lenses, which were designed and manufactured for the NM. As it was mentioned above, the effective length of lenses constitutes 50 mm and the radius of the aperture is 6,5 mm. The poles of lenses are manufactured from a high quality iron-cobalt alloy 49КФ with saturation field reaching 2.4 – 2.5 T. The tips of poles have cylindrical shape (the cylinder radius is 6.5×1.12=7,28 mm according to the recommendations given in ref. [15], the width of cylindrical part of a pole equals to10.29 mm). The total of pole width is 20 mm. The cylindrical surface is ganged to the lateral face of a pole by a plane, the corner between this plane and the pole axis constitutes 30^0. The end faces of poles are flat. The number of turns in each of 4 pole windings are 380.

The lens yoke is manufactured from a high quality iron and it has the shape of an empty cylinder (the external radius - 240 mm, the internal radius - 220 mm, the height - 55 mm). Each lens is mounted at the tuning table which allows to displace the lens in horizontal and vertical directions with a step of 10 μm and to rotate the lens around of the longitudinal axis with a step of 0.03 mrad.

Unfortunately, manufacturing the NM units coincided in time with an essential cutting of financing for this project. Nevertheless, the activities aimed at developing the NM in NSC KIPT were not stopped completely. It was decided to create the non-stationary version of NM, which could be installed at ESU-4,5 accelerator for the period of performing measurements and removed after their accomplishment.

In this version the focusing system developed for the NM is used. The beam line, collimator, the system of beam current monitoring, the vacuum system and the irradiation chamber of the existing NAM facility are also used (see Fig. 2).
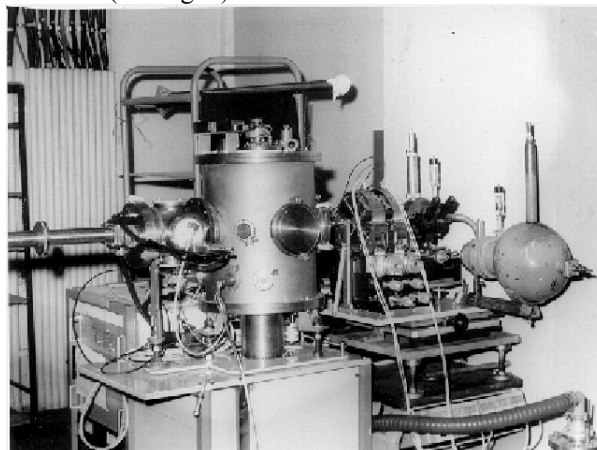

Fig.2. The general view of NM.

The short ion beam line of 12 mm diameter placed between poles of the doublet lenses connects the NAM chamber to the NM target chamber (the NM chamber incorporates a goniometer with three translation and one rotation degrees). The goniometer can move a sample with ~1 μm step. The doublet and the target chamber are mounted on the alignment table. The beam and a sample are visually observed through the window by means of the optical microscope with magnification x100. The nuclear analysis methods PIXE, PIGE, NRA, RBS are used. Parameters of NM are presented in the Table 1.

A series of analytical investigations, using this facility, was carried out. In particular, an impurity composition of synthetic diamonds (200-330 microns) was defined [23], the investigations of some composite materials were also carried out.

Table 1

Distance between object diaphragm and 1-st lens of doublet, mm	3310
Distance between lenses, mm	50
Distance between 2-nd lens and target, mm	165
Effective length of lens, mm	50
Excitation of 1-st lens (φ_1)	0,58987
Excitation of 2-nd lens (φ_2)	0,72285
Demagnification coefficient in horizontal plane (M_x^{-1})	6,6
Demagnification coefficient in vertical plane (M_y^{-1})	30,9
Beam sizes of microprobe, μm^2	5×3
Proton energy, MeV	2,4
Maximal proton energy, MeV	3,5

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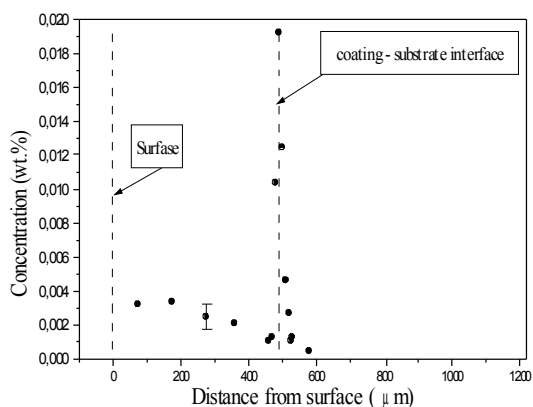*Серия:* Ядерно-физические исследования (35), с. 98-101.

98

Fig.3.The depth profile of fluorine in *W CVD* coating.

For example, the depth profile of fluorine in *W* CVD coating on the *W* substrate is presented in fig.3. This profile was obtained by PIGE technique using the reaction $^{19}F(p,p'\gamma)$. In the region of coating-substrate boundary an increase in fluorine concentration one order of magnitude is observed. Such information can be useful for technology perfecting, because often fluorine can be a harm impurity.

The NM of NSC KIPT is the first and the only one in Ukraine. Now by means of NM the analytical investigations are carried out in cooperation with NSC department of solid state physics and materials and other organizations.

The work on further improving the NM parameters and perfecting its systems will be prolonged. Now the plan of the arrangement of the stationary NM is considered.

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