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On nanoscale phenomena in the electroacoustic sputtering process*

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К вопросу о наноразмерных явлениях в процессе электроакустического напыления***

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Introduction. The effect of variable parameters of the electroacoustic sputtering (ELAS) process on the characteristics of the crystalline structure of hardening coatings is studied. The ELAS parameter values providing nanostructured cover coatings for machine parts and cutting tools are determined. Hardening through using such coatings allows achieving a significant (5-10 times) increase in the life of hardenable machine parts and various tools designed for mechanical processing. To obtain coatings with the desired properties of the surface layer, nanocrystalline materials should be selected. In this case, a certain content of the amorphous phase is permissible.
Materials and Methods. To carry out the X-ray structural analysis, the X-ray diffraction Russian-made device DRON-3M was used. The Scherrer-Wilson method was applied to determine the granularity of particle blocks from the value of the intrinsic broadening of the diffractogram peaks. The conclusions obtained in this paper are based on the method of separation of the affecting factor contributions into broadening the diffraction reflection peaks (the Warren-Averbach method).
Research Results. Depending on the process conditions and the technique for obtaining nanostructured materials, a non-uniquely interpretable change in the indices of the diffraction peaks broadening occurs, which is generally characteristic of nanocrystalline metals. One of the possible explanations for this phenomenon is the presence of a nanosized effect in the hardened layer. The occurrence of the nanocrystalline structure in the sputtered layer verifies the calculated values of the dimensions of the coherent scattering regions (CSR). The occurrence of affecting values of the misorientation angle of the crystal structure is verified by the CSR value for the investigated 110 and 220 reflexes, which is supported by a high percentage of the amorphous phase.

Введение. Исследовано действие варьируемых параметров процесса электроакустического напыления (ЭЛАН) на характеристики кристаллической структуры упрочняющих покрытий. Выявлены значения указанных параметров, обеспечивающие получение наноструктурных защитных покрытий деталей машин и режущего инструмента. Упрочнение при помощи подобных покрытий позволяет достичь значительного (в 5–10 раз) повышения ресурса работы упрочняемых деталей и инструментов, предназначенных для механообработки. Для получения покрытий с заданными свойствами поверхностного слоя следует выбирать нанокристаллические материалы. При этом допустимо определенное содержание аморфной фазы.
Материалы и методы. Использован рентгеноструктурный анализ, который проводился на рентгеновском дифракционном аппарате отечественного производства «ДРОН-3М». Метод Шеррера — Вилсона применен с целью определения зернистости блоков частиц по значению физического уширения пиков дифрактограммы. Выводы по результатам работы основаны на методике разделения вкладов значащих факторов в уширение пиков дифракционных отражений (метод Уоррена — Авербаха).
Результаты исследования. В зависимости от технологических режимов и метода получения наноструктурных материалов происходит неоднозначно интерпретируемое изменение показателей уширения дифракционных пиков, что, в общем, характерно для нанокристаллических металлов. Это явление можно объяснить, в частности, наличием наноразмерного эффекта в упрочненном слое. Присутствие нанокристаллической структуры в напыленном слое напрямую подтверждается вычисленными значениями размеров областей когерентного рассеивания (ОКР). Наличие значимых величин угла разориентирования структуры кристаллов подтверждается величиной ОКР для исследованных рефлексов 110 и 220, что подкрепляется высоким процентным содержанием аморфной фазы.

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Discussion and Conclusions. The electroacoustic scattering method is promising for obtaining nanocrystalline structures in the surface and subsurface layers of the sprayed samples. The ELAS process variables variation leads to the parameter spread of the crystal lattice and coherent scattering areas. In this case, there is no definite trend. In the future it is expected to solve the given problem. First, experiments will be conducted to determine the optimal sputtering regimes that could stimulate the formation of nanocrystalline structures. Secondly, visual observation and evaluation of the sprayed layer structure using electron microscopy is planned.

Обсуждение и заключения. Метод электроакустического напыления перспективен для получения нанокристаллических структур в поверхностном и приповерхностном слоях напыленных образцов. Изменение технологических параметров ЭЛАН приводит к разбросу значений параметров кристаллической решетки и областей когерентного рассеивания. В этом случае не выявляется определенная тенденция. В дальнейшем предполагается решение данной проблемы. Во-первых, будут проведены эксперименты с целью определения оптимальных режимов напыления, способствующих образованию нанокристаллических структур. Во-вторых, планируется визуальное наблюдение и оценка структуры напыляемого слоя при помощи электронной микроскопии.

Keywords: nanocrystalline structures, hardening coatings, crystal lattice, diffractometric studies, electroacoustic sputtering.

Ключевые слова: нанокристаллические структуры, упрочняющие покрытия, кристаллическая решетка, дифрактометрические исследования, электроакустическое напыление.

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Introduction. To obtain nanostructured hardening coatings is an urgent and long-run engineering challenge [1, 2]. Numerous studies in the field of materials science have shown that a tangible (by several times) change in strength, hardness, and wear resistance of materials is possible with a decrease in crystal grains to a certain value.

Under this investigation, it is expected to study the crystal microstructure of hardening coatings using the method of electroacoustic sputtering (ELAS) [3, 4, 5]. Such hardening involves the use of an electric spark (highly-concentrated power flotation) and ultrasonic longitudinally-torsional vibrations. ELAS-hardening, using a specialized installation, enables to increase the operational life of parts and tools by 5-10 times.

In the paper, the ELAS operational parameters vary in order to identify their values, which contribute to obtaining nanostructural protective coatings for the parts and cutting tools. To create protective coatings with the required properties, alloys in the amorphous state are often used. However, in certain areas, nanocrystalline materials are used more often than the amorphous ones. Nanocrystals relax to a far lesser extent in use of the coating, and they are highly competitive with amorphous materials in their properties. Obviously, to obtain coatings with the desired and stable properties of the surface layer, nanocrystalline materials should be selected, though with a certain content of the amorphous phase.

Materials and Methods. Resonance methods, X-ray diffraction analysis, electron microscopy, and some other techniques [2] are widely used to identify nanocrystalline materials in the surface layer. In the present work, the most accessible method based on identifying the diffraction reflection broadening under the X-ray structural analysis of samples is used to evaluate the microstructure parameters. “DRON-3M”, the X-ray diffraction Russian-made apparatus, was used for X-ray diffraction analysis. The characteristic radiation of the iron anode with the release of λK_{α} spectral line of the sample, as well as of the detector, was analyzed. The X-ray tube specifications were 25 kV, 5 mA. The Bragg - Brentano method of radiation focusing was used [6]. The selected detector parameters are as follows: motion speed is 1 deg/min; integration time is 5s. When determining the distances between the measurement planes, the error was ± 0.001 Å, which is similar to determining the position of diffraction maxima accurate to ± 0.02 degrees. Samples of a cylindrical shape made of 45 steel were studied. The diameter and height of the sample was 6 mm.

To identify samples, special marks were applied using ELAS on the front surface of each of them. The sample without sputtering made from 45 steel was chosen as a reference one, not the sample that is close to pure α-Fe.

Research Results. The profiles obtained during the work are presented in Fig. 1.

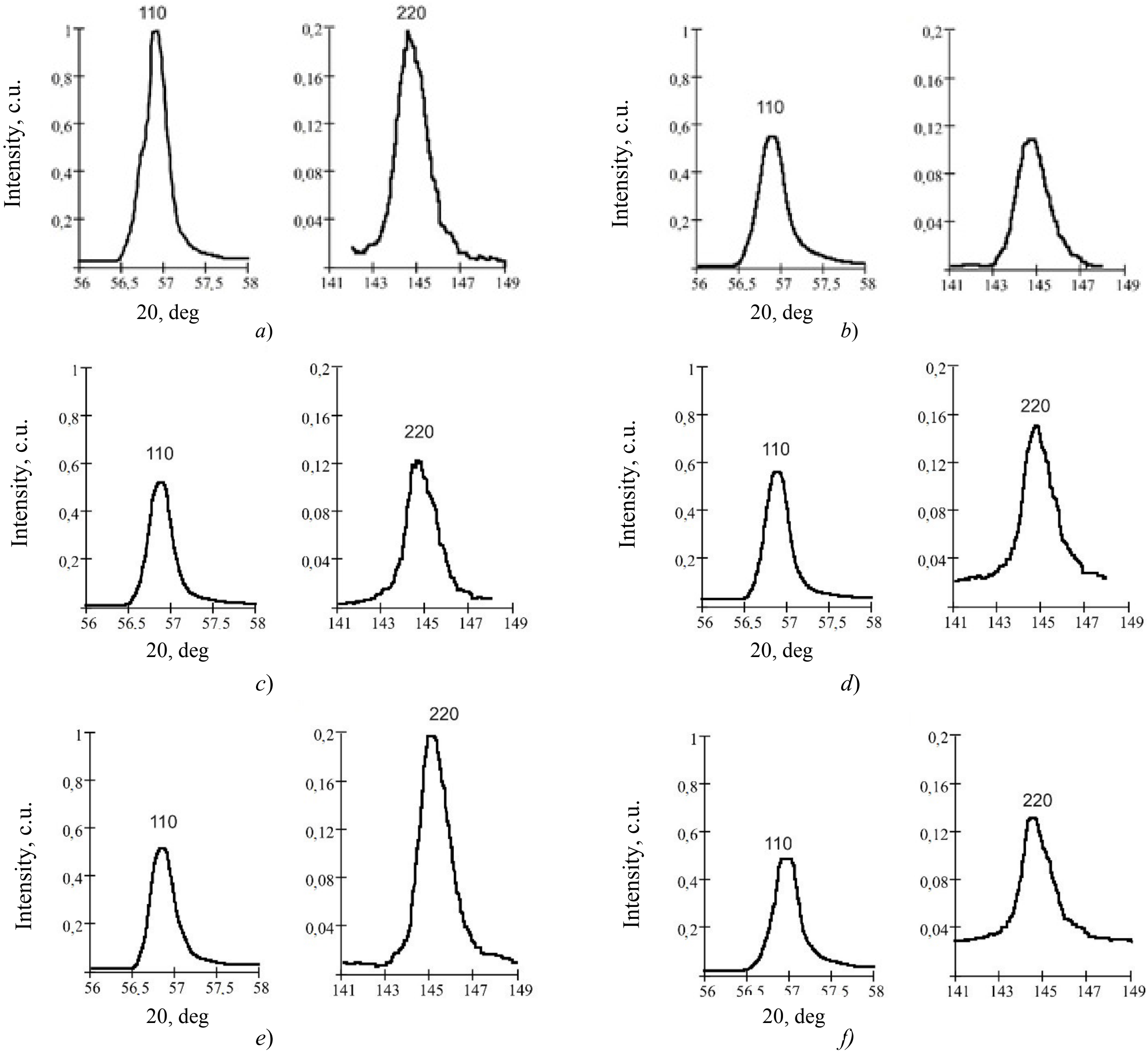


Fig. 1. Diffraction reflections with profiles: reference with coarse grain (*a*); sample for sputtering (U = 16.9 V, A = 4.9 microns) (*b*); U = 13.1 V, A = 15.2 μm (*c*); U = 12.9 V, A = 9.9 μm (*d*); U = 12.9 V, A = 5.1 μm (*e*); U = 9.1 V, A = 15.1 μm (*f*)

The shape of the diffraction profiles for the reference sample is shown in Fig. 1 (*a*), for samples with spread sputtered coating on the above modes – in Fig. 1 (*b*) - (*f*). ELAS variables are electrode-anode voltage (U) and amplitude value (A) of ultrasonic frequency oscillations.

Diffractograms of the reference and study sample show the intensity values ratio. The relationship of the crystallized and amorphous material was calculated from these data. The contribution of the texture component was not considered.

Analysis of the experimental data allows us to conclude that the main phase in most of the samples under study is α - Fe. The Scherrer-Wilson method (the best in this case) was used to determine the grain coarseness of blocks of particles according to the value of the physical broadening of the diffractogram peaks [7].

Table 1 presents the summarized data on the research results:

- β physical broadening,
- *d*-space,
- *a* dimensional parameter of the crystal lattice,
- size of coherent scattering regions.

Table 1

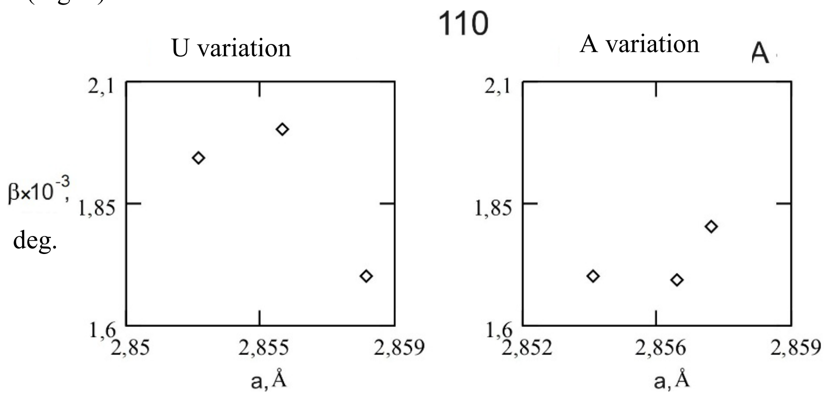
Design parameters of crystal structure and diffractogram data

Parameter / Sample	Reflex											
	110						220					
	1	2	3	4	5	эт.	1	2	3	4	5	ref.
$\beta \cdot 10^{-3}$, deg.	2.01	1.71	1.68	1.81	1.95	1.39	8.47	8.34	8.62	8.87	10.01	8.06
2θ, deg.	56.916	56.86	56.926	56.876	56.976	57.2	144.66	144.7	144.78	144.91	144.53	144.4
d, Å	2.026	2.028	2.025	2.027	2.024	2.019	1.013	1.0129	1.0126	1.0122	1.0132	1.0129
a, Å	2.8553	2.8581	2.8539	2.8567	2.8525	2.8465	2.8539	2.8561	2.8553	2.8793	2.8571	2.8571
D, nm	111	130	131	123	114	–	76	77	75	73	64	–
Amorphous component content, %	59	55	57.5	53.5	51	–	59	75	81	98	79	–

110 and 220 reflexes were selected as the basic ones.

Table 1 data demonstrate significant broadening of the diffraction peaks. It is logical to assume that such a width in our case is, to a greater extent, due to the highly dispersed structure of the crystallites than to the dislocation microstresses of the surface layer. To confirm this conclusion, the contributions from these factors are estimated by the Warren – Averbach method [8]. The center-of-gravity shift of the peaks on the diffractograms with a simultaneous increase in *a* lattice parameter to 1.15–1.17% serves as an indirect proof for the above formulated hypothesis. The shift itself is small, but meaningful (expressed in hundredths of a degree).

Consider the phenomena that occur when changing the values of the process conditions (electrical voltage and amplitude of oscillations), and their effect on the broadening value of the diffractogram peaks depending on the lattice dimensional parameter (Fig. 2).



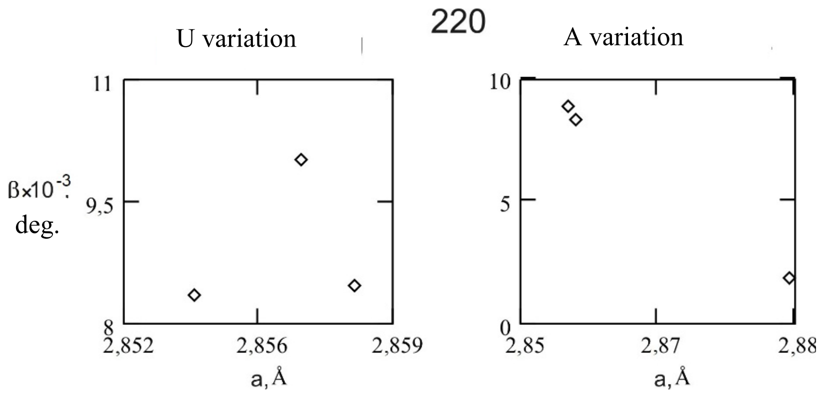
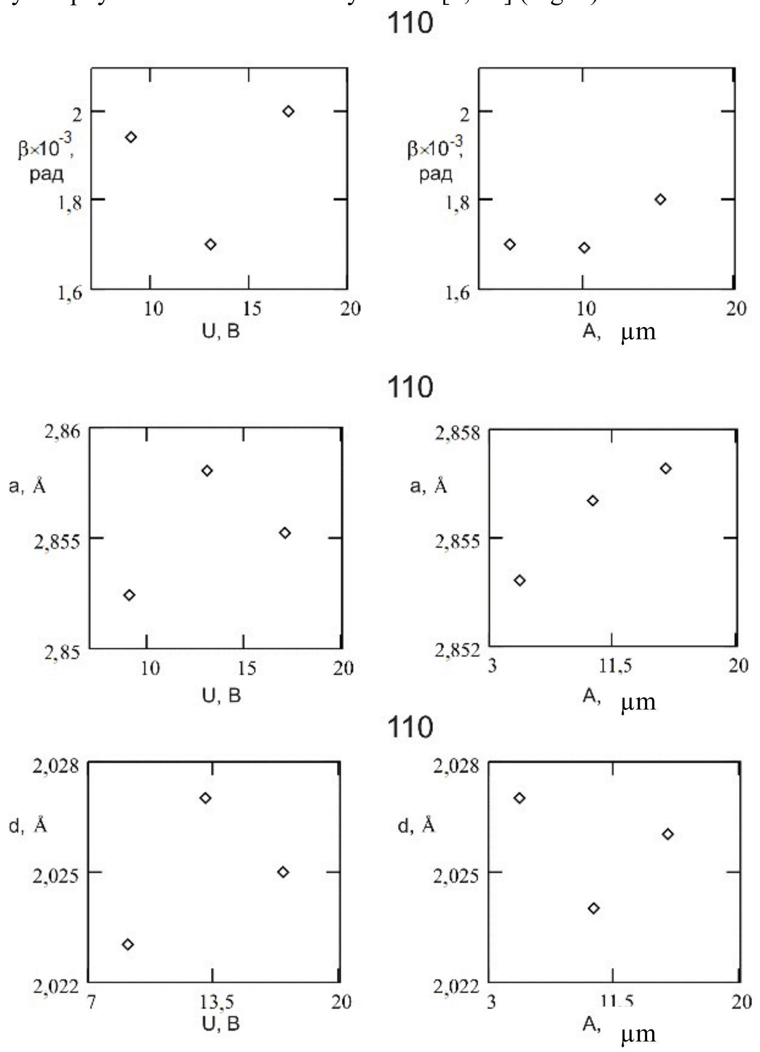


Fig. 2. Effect of ELAS regimes variation on *a* lattice parameter depending on β value of physical broadening of diffractogram peaks

As expected, a change in the deposition modes affects *a* lattice parameter without a definite traceable dependence. In this case, *a* value variations are directly proportional to the broadening value of the lines of the diffractogram lines. In [9], comparable proportional dependences can be observed for various oxides.

Such ambiguously interpolated dependences are characteristic of nanostructured materials (in particular, metals) and are determined by the physical method of their synthesis [1, 10] (Fig. 3).



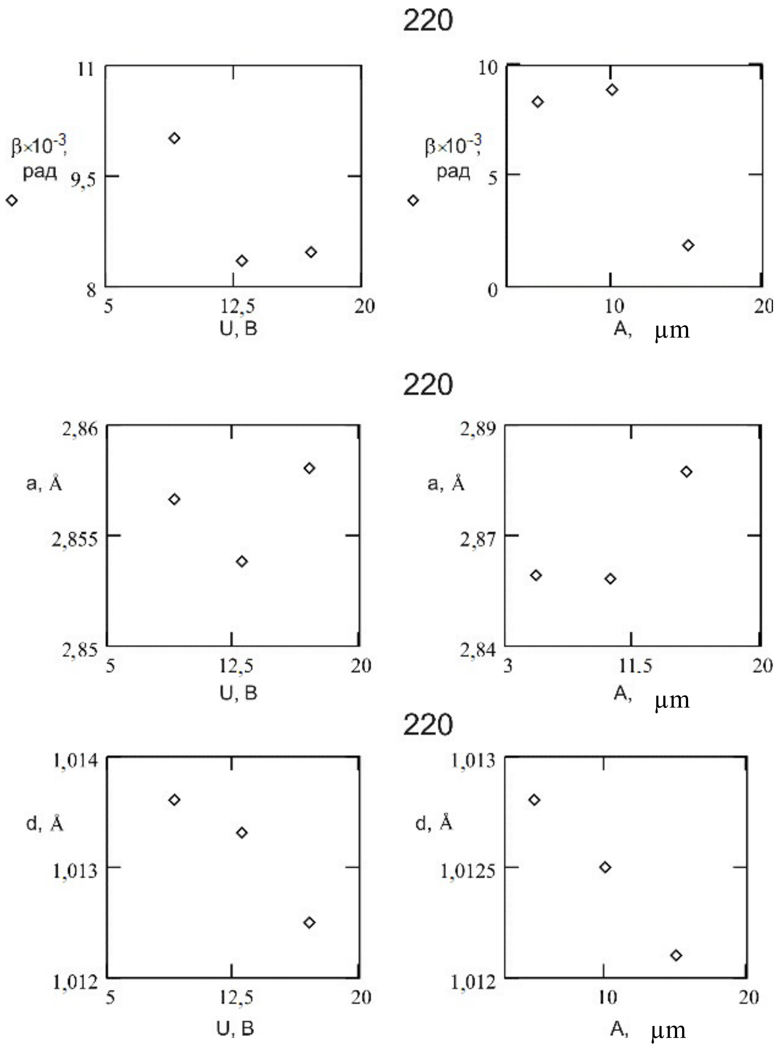


Fig. 3. Effect of ELAS variables on microstructure parameters

Perhaps, this phenomenon is explained by the occurrence of a nanoscale effect in the hardened layer. The presence of a nanocrystalline structure in the sputtered layer is directly confirmed by the calculated values of the sizes of coherent scattering regions (CSR). The significant values of the disorientation angle of the crystal structure is verified by the CSR values for 110 studied reflex, and also, noticeably, for 220 reflex, which is supported by a high percentage content of the amorphous phase. Similar phenomena can be also caused by high values of disorientation angles.

Summing up, it can be stated that all of the above phenomena (including sizes of the coherent scattering regions) directly confirm the presence of a nanoscale effect [1].

Discussion and Conclusions. Thus, the method of electroacoustic sputtering is promising for obtaining nanocrystalline structures in the surface and near-surface layers of sputtered samples. As stated in the paper, ELAS process parameter variation brings into existence the lattice parameter spread and coherent scattering regions without a definite identified tendency. In the future, this problem will be solved. This can be facilitated by the experimental determination of optimal sputtering regimes that ensure the formation of nanocrystalline structures. In addition, the structure of the sputtered layer can be observed and evaluated using electron microscopy.

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