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DETERMINATION OF THE INFLUENCE OF THE PHASE COMPOSITION OF Cu-Bi COATINGS ON THE EFFICIENCY OF SHIELDING FROM IONIZING RADIATION.

Kadyrzhanov K.K.¹, Shlimas D.I.¹, Kaniukov E.Yu.², Kaliyekperov M.E.¹

¹L.N. Gumilyov Eurasian National University, Nur-Sultan, Kazakhstan, <u>kayrat.kadyrzhanov@mail.ru</u> ²Scientific-Practical Materials Research Centre of National Academy of Sciences of Belarus, Minsk,

Belarus

The work is devoted to the study of the efficiency of shielding the radiation exposure of heavy ions depending on the phase composition of Cu-Bi coatings. As a method of producing protective coatings, an electrochemical deposition method was used. The change in phase composition was carried out by varying the applied potentials difference during deposition. The study found that the change in phase composition leads to an increase in coating density, as a result of the dominance of the CuBi₂O₄ phase and an increase in the structural ordering degree. During shielding efficiency tests, it was found that the greatest decrease in the degree of crystallinity is observed for copper coatings, for which the amorphization of the structure at the maximum irradiation fluence didn't exceed 12 %, while for coatings based on CuBi₂O₄ the value of amorphization was no more than 1.2 % of initial value. Amorphization and fragmentation of grains at high radiation doses leads to a decrease in the shielding efficiency, as well as an increase in the value of the deviation ΔU .

Keywords: protective coatings, shielding, ionizing radiation, heavy ions, radiation resistance.

Introduction

The rapid development of technologies in the field of nuclear energy and reactor building is inextricably linked to the problem of radiation safety and protection against the negative impact of ionizing radiation [1-3]. In the transition to the widespread use of microelectronic devices, as well as the reduction in the size of semiconductor chips, their resistance to the destabilizing effects of ionizing radiation plays a large role. Reduction of mass-dimensional dimensions of microelectronic devices led to a sharp decrease in the level of natural protection against ionizing radiation [4, 5]. At the same time, a negative impact that leads to destabilization of performance and failure can lead to both long-term irradiation and a single cascade of secondary dislodged particles caused by the collision of a heavy ion [6-10]. Radiation-resistant materials based on nitrides [11-13], carbides [14, 15] or nanostructured films and coatings are used as the most common methods of protection against ionizing radiation.

The most promising solution to the problem of maintaining the stability of microelectronic devices operating in conditions of high radiation background is the use of composite protective coatings of complex composition [16-20]. The choice of protective coatings is based on the principles of preservation of physical, chemical and mechanical properties as a result of prolonged exposure to external factors and loads [21-24]. Moreover, it should be noted that in most cases it is impossible to create a protective coating capable of protecting against all types of exposure, but at the same time it is possible to significantly reduce one or another type of radiation that can cause the greatest negative impact on performance [25-27].

The purpose of this work is to study the possibility of using Cu-Bi films for protection against heavy ions, as well as the effect of the phase composition on the efficiency of shielding and weakening the negative impact on microelectronic circuits. Copper-bismuth (Cu-Bi) - based coatings were selected as research objects to determine the effect of the crystal structure, including the phase composition of the synthesized coatings, as well as surface morphology (grain size) on the shielding efficiency [28,29]. The choice of this type of coating is due to the possibility of obtaining coatings with different phase compositions, including both two-phase and single-phase coatings. The change in phase composition, and hence crystal structure, can have a significant impact on shielding efficiency by varying coating density as well as absorption capacity

and affecting energy losses. The change in grain size has a great effect on dislocation density, which can also lead to strengthening of coatings and reduction of radiation damage.

1. Experimental part

As a method of obtaining protective coatings, the method of electrochemical deposition from electrolyte solutions was chosen: $CuSO_4 \cdot 5H_2O$ (238 g/l), $Bi_2(SO_4)_3$ (10 g/l), H_2SO_4 (21 g/l) [28,29]. The applied potentials difference for deposition was varied from 1.5 V to 2.0 V. The change in the applied potentials difference is due to the possibility of varying the phase composition of the synthesized coatings.

Evaluation of structural changes and phase composition of the synthesized samples was carried out using the method of X-ray phase analysis performed on a D8 Advance ECO X-ray diffractometer.

 Xe^{22+} ions with an energy of 230 MeV and fluences of $10^{13} - 10^{15}$ ions/cm² were selected for shielding efficiency tests. The irradiation was performed on a DC-60 heavy ion accelerator [12]. The choice of fluences is due to the possibility of modeling the effects of overlapping defect areas that occur during irradiation. With the selected energy of incident ions, the path length in the selected materials for testing exceeds 12-20 µm, depending on the type of ions, comparable to the thickness of the films.

The efficiency of shielding radiation damage caused by ionizing radiation will be assessed according to the method of determining the preservation of the performance of microcircuits by measuring deviations in volt-ampere characteristics before and after irradiation. The threshold value ΔU , which is characteristic of critical malfunctions of the microcircuits, will be selected as 0.1 V. Deviation of the volt-ampere characteristic above the threshold value leads to deterioration of the microcircuit and its further failure.

2. Results and discussion

Table 1 shows the initial data of the test coatings that were used to test the absorption efficiency. The thickness of the protective coatings was 10 μ m. As can be seen from the data presented in Table 1, by changing the synthesis conditions, in particular the applied potential difference, it is possible to control the phase composition of the synthesized coatings to obtain both single-phase coatings consisting of copper or CuBi₂O₄ and a mixed composition. At the same time, the change in phase composition leads to an increase in the density of the coatings, as a result of the dominance of the CuBi₂O₄ phase and an increase in the degree of structural ordering (degree of crystallinity). However, in the case where the CuBi₂O₄ phases in the coating composition become larger, an increase in grain size is observed, and thereby a decrease in dislocation density.

| Conditions for obtaining | Phase composition, phase ratio | Average crystallite size, nm | Density, g/cm ³ | Crystallinity degree, % | Dislocation density, 10 ¹¹ cm ⁻² |
|--------------------------|---|------------------------------|-------------------------------|----------------------------|--|
| 1.5 V | Cu – FCC – 100 % | 73.5 | 9.17 | 81.2 | 0.185 |
| 1.6 V | $\begin{array}{c} Cu - FCC - 75 \ \% \\ CuBi_2O_4 - HCP - \\ 25 \ \% \end{array}$ | 78.3 | 11.42 | 85.3 | 0.163 |
| 1.75 V | $\begin{array}{c} Cu - FCC - 25 \ \% \\ CuBi_2O_4 - HCP - \\ 75 \ \% \end{array}$ | 84.5 | 12.44 | 88.3 | 0.141 |
| 2.0 V | CuBi ₂ O ₄ – HCP – 100 % | 96.3 | 12.48 | 89.7 | 0.108 |

Table 1 - Initial sample data.

To assess the effect of phase composition and crystal structure on shielding efficiency, Xe^{22+} ions with an energy of 230 MeV and fluences of $1x10^{13}-10^{15}$ ion/cm² were selected as ionizing radiation. Evaluation of efficiency of radiation damage shielding caused by ionizing radiation was carried out according to the method of determining serviceability of microcircuits by measuring deviations of volt-ampere characteristics before and after irradiation. The threshold value ΔU , which is characteristic of critical malfunctions of the microcircuits, was selected as 0.1 V. Deviation of the volt-ampere characteristic above the threshold value leads to deterioration of the microcircuit and its further failure. Figure 1 shows experimental data of ΔU value change depending on irradiation dose for all tested coating samples.



Fig.1. Data on changes in the value of ΔU depending on the radiation dose

As can be seen from the presented data, at a radiation dose of 1013 ion/cm², all the tested protective coatings show a high level of absorption of radiation damage, and the deviation ΔU does not exceed 0.05-0.06 V. However, an increase in the radiation dose leads to a significant change in the behavior of ΔU for the studied coatings. For coatings with a crystalline structure characteristic of copper, as well as with a large content of the Cu-FCC phase in the structure, a decrease in absorption efficiency is observed, and an increase in ΔU value that exceeds the threshold value. Further increase of irradiation dose results in deviation of ΔU value being more than 0.15-0.17 V, which indicates sharp decrease of absorption capacity of coatings and deterioration of protection degree.

For coatings dominated by the CuBi_2O_4 phase, as well as single-phase coatings based on CuBi_2O_4 , the deviation of ΔU is significantly lower than for copper coatings, indicating a higher degree of resistance to radiation damage and a high degree of absorption of radiation damage. Figure 2 shows a graph for evaluating the efficiency of reducing radiation damage at maximum irradiation fluence, which was calculated as a result of a comparative analysis with the deviation of the ΔU value for microcircuits without protective coatings.



Fig.2. Graph of radiation damage reduction efficiency assessment at maximum radiation fluence.

As can be seen from the data presented, coatings based on CuBi_2O_4 for which the value of the shielding efficiency was 6.96 are most effective. The increase in shielding efficiency for coatings is CuBi_2O_4 associated with high radiation resistance to radiation damage, as well as resistance of the crystal structure to degradation and amorphization as a result of irradiation. Figure 3 shows the data of changes in the value of

the degree of structural ordering as a result of irradiation, as well as an estimate of the change in grain sizes and dislocation density (see Figure 4).



Fig.3. Dynamics of changes in the degree of crystallinity of the coatings under study depending on the radiation dose

As can be seen from the data presented, the greatest decrease in crystallinity is observed for copper coatings, for which amorphization of the structure at maximum irradiation fluence was more than 12 %, while for coatings based on CuBi_2O_4 , the value of amorphization didn't exceed 1.2 % of the initial value. A high degree of amorphization for copper coatings leads to disordering of the crystal structure, large deformations of the crystal lattice, crushing of crystallites and an increase in dislocation density. According to the presented dependences, the change in the grain size for coatings containing CuBi2O4 with an increase in the radiation dose is less pronounced than for copper coatings. At the same time, an increase in the radiation dose leads to the processes of grain fragmentation, thereby increasing the dislocation density of defects.



Fig.4. Graphs of changes in structural parameters: a) Change of grain size depending on radiation dose; b) Change of dislocation density depending on radiation dose.

Thus, during the study, it was found that a change in the crystal structure, including the phase composition of synthesized coatings, leads to an increase in the efficiency of shielding ionizing radiation, as well as a decrease in the degree of amorphization of coatings as a result of the accumulation of radiation damage. The amorphization and crushing of grains at high radiation doses leads to a decrease in the shielding efficiency, as well as an increase in the deviation ΔU . It has been found that coatings based on CuBi₂O₄ have

the best radiation resistance to degradation and amorphization, as well as the efficiency of shielding from ionizing radiation.

Conclusion

During the study, it was found that a change in the crystal structure, including the phase composition of the synthesized coatings. This change leads to an increase in the shielding efficiency of ionizing radiation, as well as a decrease in the degree of amorphization of coatings as a result of the accumulation of radiation damage. Amorphization and fragmentation of grains at high radiation doses leads to a decrease in the shielding efficiency, as well as an increase in the value of the deviation ΔU . It was found that the best radiation resistance to degradation and amorphization, as well as the efficiency of shielding from ionizing radiation, is possessed by $CuBi_2O_4$ -based coatings. In the future, studies to compare the shielding efficiency of synthesized coatings with commercial analogues based on silicon carbide, titanium nitride, aluminum nitride and tungsten oxide will be carried out.

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