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BACK-SCATTERING AND NEUTRALIZATION COEFFICIENTS OF NITROGEN IONS ON IRON SURFACE

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The coefficients of back-scattering and neutralization of nitrogen ions (N^+) on iron surface during ion bombardment at the processes of ion sputtering and niriding have been calculated using TRIM code. The pure iron target and two-element targets, Fe+Mo (1 and 5 %), Fe+W (1 and 5 %), were tested. The ion energy was from 0.1 to 70 keV. The ions N^+ are considered to be back-scattering in the form of energetic N atoms. The calculations showed the back-scattering and neutralization coefficients are tens of percent for low energies N^+ ions and several percent for energies of tens kiloelectron-volts. The average penetration depth of the ions into the Fe target and the ion sputtering coefficients are calculated, too.

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INTRODUCTION

Plasma physics and technology deal with different kinds of gas discharges, in which various types of high-energy species, including charged and neutral particles, are generated and interacting with gas medium and solid electrodes [1, 2]. Herein, the electrodes are not only collectors of particles, but also diffuse reflectors of particles: electrons from anodes, heavy particles from cathodes. The latter particles are the former ions and neutrals bombing the cathodes in gas discharges or the targets in ion devices. In addition, it should be noted various types of secondary emission from electrodes, including sputtering and evaporation of their surface.

Of all the noted processes of interaction of heavy particles with cathodes/targets, the data on the process of back-scattering or reflection of ions are least represented in the literature. This process is more or less known for light ions [3] and for some types of ions used in back-scattering spectroscopy [3, 4]. In the literature on ion sputtering of thin films, there are some data on the back-scattering of argon ions in connection with the problem of argon incorporation into deposited films and the influence of the energy, transferred by back-scattered fast Ar atoms, on the formation of the film microstructure [2]. At the same time, there are very few data on the back-scattering of nitrogen ions, although nitrogen is widely used in the important technologies of ion implantation and nitriding, nitride coating, for sterilization as well [5-8]. It should also be noted that the bombardment of the solid surface with molecular N_2^+ ions (namely, such ions are mainly generated in discharges) leads to the dissociation of the molecules into atomic particles, which then act individually [6].

The atomic nitrogen (N or N^+) is very reactive that promotes the processes with its participation; therefore, it is important to be aware of atomic nitrogen generation by the back-scattering in the gaseous medium in the vicinity of the surface treated by ion bombardment.

The atomic and molecular ions are neutralized upon collision with metals; hence, almost all of them are back-scattered in the form of high-energy neutral atoms [3, 4]. Correspondently, the calculated back-scattering coefficients will also characterize the generation of high-energy atomic neutrals near the electrode bombarded by different nitrogen ions.

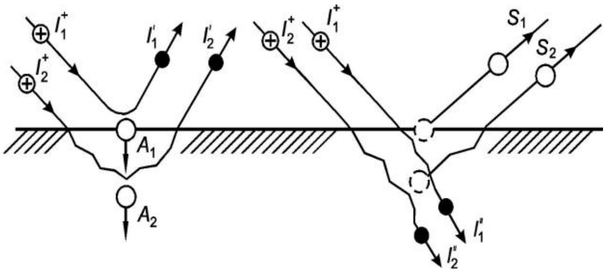
Since the technology of nitriding is aimed mostly at the processing of iron materials [8], this paper is devoted to such a case.

1. METHOD OF RESEARCH

The research was performed with the TRIM (TRansport of Ions in Matter) code, which presents a statistical simulation of ion motion within the target material using the Monte Carlo method [9, 10]. The TRIM code in comparison with the others is often discussed elsewhere in literature [5, 6, 10] and it is shown that calculations with this code give a good agreement with the experiment. Also, to be additionally sure of obtaining correct results on back-scattering, the coefficients of iron sputtering by nitrogen ions have been calculated with the same TRIM program and then compared with experiments [11].

The process of scattering the primary bombarding ions is modeled as a sequence of random binary collisions with target atoms. A random arrangement of atoms within the target, *i.e.* its amorphous microstructure, with a fixed average distance between atoms, is assumed. On a rectilinear free path, the energy of the primary particle continuously decreases by the value of electronic (inelastic) losses and after the collision by nuclear (elastic) losses with the transfer of energy to the recoil atom of the target (the Ziegler-Biersack-Littmark collision model is used [9]). The motion of recoil atoms in their multistage collisional processes is also traced. The trajectory of the primary particle (the former ion) or the recoil atom is terminated if their energy drops to a given value (~10 eV) or if they go beyond the target.

Schematically modeled processes of ion back-scattering from the target, wandering ions within the target and sputtering target atoms are shown in Figure. Primary N^+ ions (I^+) can be back-scattered both from surface target Fe atoms (A_1) in a single collision and from deep target Fe atoms (A_2) as a result of multiple collisions with the corresponding loss of initial energy or get stuck within the target. The target Fe atoms (recoil atoms), which have received sufficient kinetic energy in the direction normal to the surface as a result of collisions, form a stream of sputtered Fe atoms (S). The exit of particles from the target is possible if the energy of recoil atoms is greater than the value of the potential surface barrier or the surface binding energy (SBE). The SBE is usially determined by the sublimation energy of the target material [3, 9, 10]. Such approach is accepted in the TRIM code.



Schemes of the processes of scattering N atoms from the surface target and within the volume of the target as well as of sputtering target Fe atoms during ion bombardment. $A_{1,2}$ *are recoil Fe atoms of the target material,* $I^+_{1,2}$ *are primary ions* N^+, $I'_{1,2}$ *are energetic N atoms scattered from the target,* $I''_{1,2}$ *are energetic N atoms moving and scattered inside the target,* $S_{1,2}$ *are sputtered target Fe atoms*

Most of the particles back-scattered from the target are neutralized ions that are neutral N atoms. The main characteristics of the process of back-scattering are the integral coefficients of back-scattering (reflection) of ions/atoms R_N and energy R_E. The coefficient $R_{N.}$ is defined as the ratio of the total number of scattered atoms N_R, regardless of their energy, angle of departure and charge, to the number of primary particles N_0 (in terms of atoms) bombarding the target (or discharge cathode): $R_N = \frac{N_R}{N_0} = \frac{1}{N_0}\int_{2\pi}\int_0^{E_i} F_N \, d\varepsilon d\Omega$, where F_N is the distribution of N_R particles over the energy ε and angles of departure from the target per unit solid angle Ω, E_i is the energy of the primary ions. Coefficient $R_E = \frac{\bar{E}N_R}{E_i N_0} = \frac{1}{E_i N_0}\int_{2\pi}\int_0^{E_i} \varepsilon F_N \, d\varepsilon d\Omega$ is defined as the fraction of energy carried away by back-scattered atoms. Average energy of back-scattered atoms is $\bar{E} = \frac{R_E}{R_N}E_i$. The ion sputtering coefficient S is determined similarly: $S = \frac{N_S}{N_0} = \frac{1}{N_0}\int_{2\pi}\int_0^{E_i} F_S \, d\varepsilon d\Omega$, where N_S is the number of sputtered atoms, F_S is the distribution of N_S atoms over the energy ε and angles of departure from the target per unit solid angle Ω. The coefficients R_N, R_E, and S depend on E_i, the angle of incidence of ions, atomic numbers and masses of bombarding ions and target atoms. An important characteristic of the back-scattering atoms is their distribution over the angles of departure from the target. Experiments show that in the case of normal incidence of ions on the non-monocrystal target, the angular distribution of scattering particles obeys the cosine law [3, 4]. In the paper the normal incidence of ions on the target is assumed.

Since iron alloys in the forms of various sorts of steel are most often subjected to ion treatment [8], the effect of Mo and W additives to iron on the back-scattering of nitrogen ions was studied, too. In this case, we proceeded from the assumption that the heavier Mo and W atoms will act as scattering centers due to their larger mass with directing nitrogen particles from inside the target towards the surface. Accordingly, recoil Fe atoms will also be directed towards the surface and create a stream of sputtered material. Testing lighter additives commonly found in alloy steels is of less interest. The partial sputtering coefficients for Fe, Mo and W were calculated with the same TRIM code.

The studies were carried out in a wide range of bombarding ion energies (0.1...70.0 keV), where low energies correspond to the technology of low-voltage nitriding and ion sputtering, while high energies correspond to the nitriding by ion implantation.

In the calculations, as it is said above, the presence of the potential surface barrier, which determined by the SBE value, is used [3, 9, 10]. In Table, the SBE value for the single-element Fe target is given. In the calculations for the two-element targets, the arithmetic averages of the sums of SBE for both elements are used [9, 10], see Table, too.

2. CALCULATION RESULTS AND DISCUSSION

Table gives the results of calculations of the coefficients of ion back-scattering R_N, the average penetration depth of nitrogen ions into the target body L, the ion sputtering coefficients S for the single-element (Fe) and the partial sputtering coefficients (see numbers through a slash) for the two-element (Fe+Mo, Fe+W) targets at different energies of atomic N^+ ions, bombing the targets. The two-element targets contain one or five percent additives of Mo and W to Fe.

As mentioned above with reference to [3, 4], the ions scattered by the metal are almost completely neutralized, so we believe that the R_N coefficient also determines the ion nitrogen neutralization coefficient.

We are not aware of the experimental data on the R_N values for the combination “N^+ ion – Fe target” and we cannot directly verify the adequacy of the obtained R_N values. However, the indirect confirmation of the adequacy can be the fact that the ion sputtering coefficients calculated with the same TRIM program are quantitatively consistent with published S values for iron [11]. The extremal nature of the obtained dependence of the sputtering coefficient S on the energy of N^+ ions with a broad maximum at energy in the range of 2...5 keV is confirmed, too [11].

Coefficients of back-scattering R_N, average penetration depth of nitrogen ions into the target body L and ion sputtering coefficients S

Energy of N^+ ions (E_i), keV	Parameters	Target (cathode) material				
		Fe	Fe + 1 % Mo	Fe + 5 % Mo	Fe + 1 % W	Fe + 5 % W
		Surface binding energy, eV				
		4.34	5.585	5.585	6.51	6.51
0.1	**R_N**	**0.26**	**0.23**	**0.24**	**0.23**	**0.24**
	L, Å	7	7	7	7	7
	S	0.33	0.25/0.005	0.23/0.01	0.02/0.003	0.20/0.008
0.2	**R_N**	**0.22**	**0.21**	**0.22**	**0.22**	**0.22**
	L, Å	10	10	10	10	10
	S	0.51	0.38/0.007	0.37/0.02	0.31/0.005	0.30/0.01
0.3	**R_N**	**0.20**	**0.20**	**0.21**	**0.20**	**0.21**
	L, Å	12	12	12	12	12
	S	0.65	0.48/0.008	0.47/0.02	0.41/0.005	0.37/0.02
0.4	**R_N**	**0.19**	**0.20**	**0.19**	**0.20**	**0.20**
	L, Å	14	14	14	14	14
	S	0.77	0.56/0.01	0.54/0.02	0.46/0.007	0.44/0.02
0.5	**R_N**	**0.16**	**0.18**	**0.19**	**0.19**	**0.19**
	L, Å	16	16	16	16	17
	S	0.83	0.62/0.01	0.60/0.02	0.54/0.007	0.50/0.02
0.6	**R_N**	**0.18**	**0.18**	**0.18**	**0.19**	**0.19**
	L, Å	18	18	18	18	18
	S	0.92	0.66/0.01	0.64/0.03	0.55/0.01	0.56/0.02
0.7	**R_N**	**0.17**	**0.18**	**0.17**	**0.18**	**0.18**
	L, Å	19	20	20	20	20
	S	0.97	0.70/0.01	0.67/0.03	0.60/0.01	0.57/0.02
0.8	**R_N**	**0.17**	**0.17**	**0.17**	**0.17**	**0.18**
	L, Å	21	21	22	22	22
	S	1.01	0.72/0.01	0.71/0.03	0.61/0.01	0.60/0.02
0.9	**R_N**	**0.17**	**0.17**	**0.17**	**0.17**	**0.18**
	L, Å	23	23	23	23	24
	S	1.04	0.77/0.01	0.74/0.03	0.63/0.01	0.63/0.02
1.0	**R_N**	**0.16**	**0.16**	**0.16**	**0.17**	**0.18**
	L, Å	25	25	25	25	25
	S	1.06	0.78/0.01	0.74/0.03	0.66/0.01	0.66/0.02
2.0	**R_N**	**0.14**	**0.14**	**0.15**	**0.15**	**0.16**
	L, Å	40	39	40	39	40
	S	1.20	0.91/0.01	0.91/0.04	0.76/0.01	0.75/0.03
5.0	**R_N**	**0.11**	**0.12**	**0.12**	**0.12**	**0.13**
	L, Å	77	78	77	78	79
	S	1.18	0.89/0.02	0.92/0.04	0.81/0.01	0.76/0.03
10.0	**R_N**	**0.09**	**0.09**	**0.09**	**0.1**	**0.1**
	L, Å	135	135	136	135	138
	S	1.09	0.83/0.01	0.80/0.04	0.71/0.01	0.72/0.03
20.0	**R_N**	**0.07**	**0.07**	**0.07**	**0.07**	**0.08**
	L, Å	244	247	249	251	249
	S	0.87	0.72/0.01	0.63/0.03	0.54/0.01	0.55/0.02
40.0	**R_N**	**0.04**	**0.04**	**0.04**	**0.05**	**0.05**
	L, Å	461	459	462	459	462
	S	0.63	0.48/0.01	0.45/0.02	0.42/0.006	0.41/0.02
70.0	**R_N**	**0.03**	**0.03**	**0.03**	**0.03**	**0.04**
	L, Å	773	779	775	781	776
	S	0.46	0.35/0.006	031/0.01	0.28/0.004	0.29/0.01

The decreasing character of energy dependence of the R_N coefficient for $N^+$is consistent with the data for other ions [3, 12]. The obtained data show that at ion energies $E_i \leq 100$ eV, which are typical for nitriding at pressures about 100 Pa, more than a quarter of the bombarding ions are back-scattered in the neutral state.

And the higher the pressure near the target (cathode), the more back-scattered nitrogen atoms is to be, since the energy of the bombarding ions decreases due to their often collisions with gas molecules. At ion energy of hundreds of electron-volts, the R_N coefficient is 16...20 %, at energy of several kiloelectron-volts – about 10%, at energy of tens of kiloelectron-volts – a few percent.

In most of our cases, the energy of back-scattered N atoms is tens percent of E_i but the R_E value is less than the R_N value by about an order of magnitude as with other light gas ions [3, 12].

At energy of up to 200 eV, the average depth of ion ballistic penetration into iron and its alloys is several angstroms; at energy up to 10 keV – tens of angstroms and hundreds of angstroms at higher energies.

The analysis of changes in the values of the calculated parameters makes it possible to assess how heavy metal atoms in iron act as scattering centers. In general, the addition of W, as the heaviest element, has a noticeably stronger effect on the change in parameters compared to the addition of Mo. The influence of the additives on different parameters manifests itself in different ways. The R_N increases with increasing additives; at low energies the increase of R_N for Fe + 5 % W approximately corresponds to the percentage of W, *i.e.* ~ 5 %; at high energies, the effect is greater (10...20 %). The additives have little effect on the average depth of penetration of bombarding particles into the target (at the percentage level at energies of tens of kiloelectron-volts); at low energies the effect is not noticeable. The sputtering of iron decreases with increasing addition of both Mo and W.

The noted features can be explained by the opposite scattering vector of nitrogen ions and recoil atoms in collisions with Fe, Mo and W atoms. So, when nitrogen ions and recoil atoms move down into the target, they can be scattered by heavy atoms towards the surface; when nitrogen ions and recoil atoms move upwards, scattering downwards is possible. The scattering downwards decreases ion sputtering coefficients.

CONCLUSIONS

Thus, kinetic simulation by the Monte-Carlo method of the processes of interaction of nitrogen ions (N^+) with iron surface was successfully fulfilled. The ion energy was 0.1...70 keV; the materials of the target (it may be a discharge cathode) were pure iron (Fe) and iron with addition of Mo (1 or 5 %) and W (1 or 5 %). The coefficients of back-scattering of ions N^+ as neutral N atoms and ion sputtering of the iron targets, as well as average penetration depth of nitrogen ions into the target body, have been calculated.

The results obtained on the back-scattering of N^+ ions do not contradict the data for other types of ions, and the values of the coefficient of ion sputtering of iron are consistent with the experiment from the literature, which indicates the adequacy of the calculation results.

It is established that the back-scattering coefficient of the bombarding ions was rather high, more than 25 % at ion energies $E_i \le 100$ eV, and about 10 % at ion energies of several kiloelectron-volts.

Since positive ions are neutralized upon contact with the metal due to the Auger process, the back-scattering coefficient and the ion sputtering coefficient are practically independent of what the particle bombards the metal, that is charged or neutral ones. In this regard, the results of calculations can also be applied to the bombardment of Fe by high-energy nitrogen atoms (N).

This fact, taking into account the dissociation of molecular nitrogen ions during metal bombardment, makes it possible to apply the results of calculations of the back-scattering and ion sputtering coefficients to the case of bombardment of iron by molecular ions N_2^+. For this, the values of the calculated coefficients for the half energy of the bombarding ions N_2^+ (*i.e.* per one particle in the molecule) are used, followed by their doubling.

The same can be done in the case of bombardment of iron with high-energy molecules N_2. Such molecules are generated during the charge exchange of ions N_2^+ on neutral nitrogen molecules N_2, when the ions N_2^+ move towards the target or cathode in a gaseous medium.

The obtained data on the back-scattering of nitrogen ions and generation of energetic neutral atoms can be used for understanding various features of technological systems with nitrogen ion beams or plasma.

As a future task of research, it is possible to propose the study of the effect on back-scattering nitrogen ions of the creation of iron nitride inside the target.

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КОЕФІЦІЄНТИ ЗВОРОТНОГО РОЗСІЮВАННЯ ТА НЕЙТРАЛІЗАЦІЇ ІОНІВ АЗОТУ НА ПОВЕРХНІ ЗАЛІЗА

А. Кузьмичєв, М. Мельниченко

Коефіцієнти зворотного розсіювання та нейтралізації іонів азоту на поверхні заліза під час іонного бомбардування у процесах іонного розпилення та азотування були розраховані за допомогою програми TRIM. Випробовували мішень із чистого заліза та двоелементні мішені Fe+Mo (1 та 5 %), Fe+W (1 та 5 %). Енергія іонів становила від 0,1 до 70 кеВ. Вважалося, що іони N^+ розсіюються назад у формі енергетичних атомів N. Розрахунки показують, що коефіцієнти зворотного розсіювання та нейтралізації складають десятки відсотків для низькоенергетичних іонів N^+ і кілька відсотків для енергій у десятки кілоелектронвольт. Також розраховано середню глибину проникнення іонів азоту в Fe-мішень та коефіцієнти іонного розпилення.