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An algorithm for the calculation of heavy ion ranges in SiO_2

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Abstract The heavy ion ranges in amorphous SiO_2 have been calculated by using a technique based on solution of first order ODE's. Br, Au, Hg, Bi, projectiles have been chosen as incident ion. Since the target is assumed to be amorphous, Bragg's rule can be used to calculate electronic and nuclear stopping powers in the compound. Numerical solutions have been performed by using Fehlberg fourth-fifth order Runge–Kutta method. The results are compared with experimental data, as well as with the result of the Monte Carlo program SRIM and other standard procedures such as PRAL and WS [19]. It is found that the agreement between our method and the experiment is good and within 10%.

Key words ion implantation • ion range • SiO_2

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Introduction

Penetration of charged particles into solid targets has been a point of interest for various purposes such as nuclear fusion technology, materials science and radiation medicine for decades. However, this interest shifted towards the ion implantation as it becomes the major method used to form impurities into solids. It is the most frequently used technique for the doping of semiconductors in today's electronic devices. The development of basic theory of energetic particle penetration of solids is due to Bohr's pioneering classical treatment of energy loss of particles [3, 4]. Bethe [1], Firsov [8] and several others established quantum mechanics of energy loss on the basis of Bohr's concepts. An excellent review of subject is written by Sigmund [17] which summarizes earlier historical developments and importance of concept over the years.

Ion implantation is frequently used to modify surface properties of materials. Therefore, the projected range $\bar{R}_p$, as the mean ion range, is of great importance in such fields as materials science and technology, microelectronic devices and radiation medicine [17]. In device processing, dopant species are implanted into a dielectric or through a dielectric coating into underlying Si. In contrast to studies of range distributions in silicon, there are less experimental data on implantation profiles in commonly used dielectric layers such as SiO_2 [7]. Gibbons *et al.* [9] have calculated and tabulated ranges and range straggling in SiO_2 based on LSS [13] predictions. However, over the two last decade, our knowledge of stopping and theoretical approaches to the range calculations have been improved thanks to a number of publications [2, 23]. In the present work, a new technique has been applied to calculate the heavy ion ranges in amorphous SiO_2. The technique is extension of Kabadayi's method used for mono-atomic targets [11].

Our goal is to find an efficient method for easy and accurate calculation of heavy ion ranges in SiO_2. The present work essentially aims to show that by optimum choice of parameters in range calculation it is still possible to find accurate range data even in a compound target. In addition to that, in some cases the use of our technique leads to more satisfactory results than PRAL and WS concerning projected ranges of ions in diatomic targets such as SiO_2. In the present work, the following reasons make the calculations efficient and comparable to other techniques. First, our technique uses an electronic stopping power mechanism which is valid at non-relativistic velocities in all solid targets and is consistent with experimental data in a wide range of energies [13]. Primary advantage of this formula is that a single expression for electronic stopping can be used for low, medium and high energy regions. Second, an optimum choice of parameters is made, first by reducing the second order differential equation of Bowyer *et al.* [5] to the first order whereas the second order coefficients in stopping powers are considered for efficient and accurate evaluation. Since the precision of the results ultimately may depend on the numerical method employed for the ODE solution, we used a Runge–Kutta ODE solver of higher order.

Theory

There are a number of techniques using different mechanisms to calculate ranges of ions in solids. However, one of the simplest methods was improved by Biersack for slowing down of ions in matter based on the analysis of directional angular spread of ion motion as a function of energy [2]. Although this method has been widely used since 1982, it was Bowyer *et al.* who revised projected range algorithm (PRAL) due to some irregularities in finding PRAL equations. It was, then, shown that KRAL equations are superior to PRAL when they were compared with the experiment and standard theories [5]. Second order differential equation of Bowyer *et al*. for range calculation is the following

$$-\frac{Q_t}{2}\frac{d^2\overline{R}_p}{dE^2}+\left(S_t-\frac{\mu Q_n}{2E}\right)\frac{d\overline{R}_p}{dE}-\left(-\frac{\mu S_n}{2E}+\frac{(1-2\mu)Q_n}{8E^2}\right)\overline{R}_p=1. \tag{1}$$

First simplification is performed by reducing the second order differential equation into the first order. Then the equation that is required to find projected range can be written as follows if the second order energy loss moments are kept:

$$\left(S_t-\frac{\mu Q_n}{2E}\right)\frac{d\overline{R}_p}{dE}-\left(-\frac{\mu S_n}{2E}+\frac{(1-2\mu)Q_n}{8E^2}\right)\overline{R}_p=1. \tag{2}$$

Even further simplification is possible by neglecting the second order stopping parameters. Thus, the following equation can be derived

$$S_t\frac{d\overline{R}_p}{dE}+\frac{\mu S_n}{2E}\overline{R}_p=1. \tag{3}$$

In these equations, $\overline{R}_p$ stands for projected range, E is the initial ion energy and $\mu = M_2/M_1$ where M_1 is the ion mass and M_2 is the target mass. S_n and S_t stand for the nuclear stopping power and the total stopping power, respectively. Q_n is the second moment of the nuclear energy loss. In our calculations eq. (2) is used as including high order stopping inputs and being first order differential equation for fast numerical solution.

In order to solve both equations (1) or (2) numerically, coefficients of differential equation must be determined. These are mainly given by electronic energy loss, nuclear energy loss, second moment of nuclear energy loss and $\mu = M_2/M_1$. For calculating the electronic stopping power S_e, the formulas derived by Montenegro *et al*. [13] for ions moving in solid targets at non-relativistic velocities was used. These formulas differ from those used by Ziegler *et al*. [23] applied to PRAL and also from those previously used by Bowyer *et al*. [5] applied to KRAL. This formula can be applied in a wide energy range with a single expression and are easy to handle. However, Ziegler's electronic stopping power expression consists of different formulas for various energy regions and a number of fitting parameters which is time consuming process in calculation.

In calculation of stopping powers, charge state of the incident ion can be important especially for slow ions since electron capture and loss cross sections is small at higher energies. Effect of charge state of the projectile during the energy loss procedure have been studied and has extensive literature [12, 15, 16]. As the ion moves through the medium certain events such as excitation, charge exchange, ionisation occur. At high energies ionisation is the main source of energy loss, however other processes such as electron capture and loss and excitations becomes important at low energies. As the Montenegro formula combines all the probabilities from low, medium and high energy regions, it takes into account all of contributions depending on velocity of the particles. Thus, this formula can be used for all energy regions of stopping [13].

According to Bohr's stripping criteria, the particles moving faster than orbital velocity of the electrons are assumed to be stripped off its electrons. Therefore, in our calculations charge state of the incident ion is not contributing to the energy loss cascades significantly as we consider the swift ions in our calculations. Thus, this technique can be used even for the slow ions since Montenegro formula that we used in our calculations is designed for all energy regions and consider contributions from all energy loss mechanisms [13].

In the case of nuclear stopping power, we have used an expression of Ziegler *et al*. though the contribution of nuclear stopping is small at higher energies [23]. In order to obtain the projected range with high precision, it is necessary to consider higher energy loss moments in nuclear stopping. Since low energy ions are slowed down mainly by elastic collisions and lose their energy in relatively large amounts, the electronic straggling is of minor influence at low energies and contributes to range straggling only at high energy ($E >> 1$ MeV for light ions). For most applications, the second moment of the electronic energy loss

Q_e might, therefore, be neglected as in the LSS calculations [12]. The second moment Q_n of the nuclear energy loss is, however, of importance. It can be calculated using a formula given by Ziegler *et al*. [23].

Diatomic targets

In many applications of ion implantation in device processing, dopant species are implanted in dielectric layers such as SiO_2. Gibbons *et al.* [9] have calculated and tabulated the ranges and the straggling in SiO_2 based on LSS predictions. Grande *et al.* [10] have studied the projected range of Au and Bi ions in implanted amorphous SiO_2.

There are two different methods to extend our technique [11] to diatomic targets. The first method employs Bragg's rule which states that the stopping power of a compound may be calculated by the linear combination of stopping powers of the individual elements [6]. The second technique uses an averaged atomic number and an averaged atomic mass to form an artificial single element. However, Bragg's rule was applied only to the electronic stopping in PRAL. Bowyer *et al.* [5] showed that Bragg's rule is superior to the average atomic number technique. Therefore, in the present work, Bragg's rule was applied to all input quantities. Using Bragg's rule input quantities which are coefficients of differential equation (2) can be found as follows:

$$W(E) = \sum_{i=1}^{n} \left(-\frac{\mu_{[i]} S_{n[i]}(E)}{2E} + \frac{\left(1 - 2\mu_{[i]}\right) Q_{n[i]}(E)}{8E^2} \right) \tag{4}$$

$$K(E) = \sum_{i=1}^{n} \left(\frac{\left(\mu_{[i]}\right)^2 Q_{n[i]}(E)}{24E^2} \right) \tag{5}$$

$$L(E) = \sum_{i=1}^{n} \left(-\frac{\mu_{[i]} Q_{n[i]}(E)}{2E} \right) \tag{6}$$

where n is the number of elements in compounds and $W(E)$, $K(E)$ and $L(E)$ are the coefficients of the differential equations (see for details Ref. [5]).

Details of calculation

Bowyer *et al*. [5] presented a modified set of equations called the Kent optimised range algorithm (KORAL) which is designed for iterative solution. They used a novel iterative refinement technique based on the method developed by Winterbon [21] and a variable step ODE solver based on Adam's method to calculate ranges of ions in solids.

The numerical solution of eqs. (1) and (2) are, in principle, the solution of an initial value problem where the initial conditions must be well defined. For this purpose, we used the same method as that proposed by Biersack [2] in the low energy region. In a first step, our program calculates the initial parameters of electronic stopping power, nuclear stopping power and nuclear energy loss straggling. These results are then used to determine the coefficients of the differential equations at various points of the energy grid. A high order Runge–Kutta technique is applied to find numerical solution of eq. (2).

Results and discussion

In this section, a number of comparisons with experimental data for implantation of heavy ions such as Hg, Au, Bi and Br into amorphous SiO_2 were made to show the reliability of our method. Our results with respect to the projected range of Au ions implanted into amorphous SiO_2 are also compared with the results calculated from PRAL97, SRIM and LSS, with the numerical results of Wang and Shi [18] and with the experimental data taken from Grande *et al.* [10]. The atomic density of SiO_2 target was assumed to be 2.21 g/cm^3 in the present calculation. Figure 1 is a plot of projected range *vs*. incident ion energies for implantation of Au ions into amorphous SiO_2. The solid curve represents calculated results using our technique, and squares show the experimental data of [10] and a comparison with other standard methods is also given in Fig. 1. We found good agreement with experiment when compared with other methods. In Table 1, presented are results and a comparison with SRIM, PRAL97, WS, LSS and experiment for the incident ion energies between 15 keV and 400 keV for Au implantation into SiO_2. LSS data was taken from Gibbons *et al.* [9]. SRIM calculations were performed in a standard PC, the number of ions being 10^5 for each simulation and the density of SiO_2 target was 2.21g/cm^3 for all the ion target combinations.

As it is shown in Fig. 1, there is close agreement between the calculated projected ranges and the experimental data with the average error of 6.86% for the case of Au implantation into SiO_2. However, the average errors for PRAL, SRIM, LSS and WS are 7.10, 7.27, 25.62 and 11.68, respectively (see Table 2). This comparison shows that our work, SRIM and PRAL give similar results; however, LSS and WS predictions differ from experiment significantly with greater average error rates. We found good agreement with the literature even with the simplifications that we introduced in the current work. Thus, it is seen that the projected range calculated using our method yields

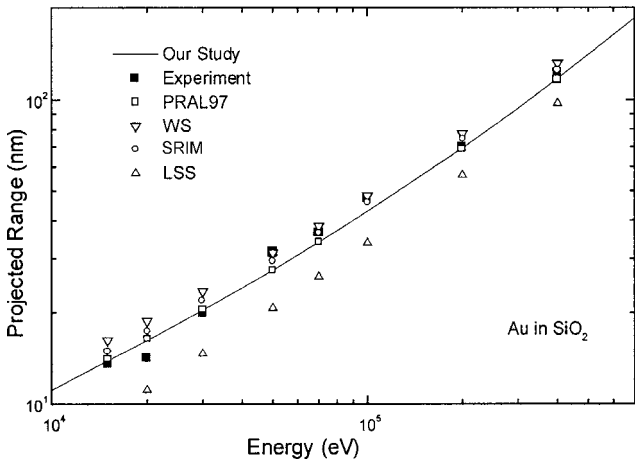

Fig. 1. Comparison of calculated projected ranges $\bar{R}_p$ of Au ions implanted into SiO_2 with PRAL, WS, SRIM, LSS and with experimental data. The solid line represents the data calculated by the present method.

Table 1. Experimental, PRAL, SRIM, LSS, WS and the calculated values of projected range for Au ions implanted into SiO_2.

Energy (keV)	Experiment (nm)	PRAL (nm)	WS [19] (nm)	SRIM (nm)	LSS (nm)	Our study (nm)
15	13.5	14.1	16.2	14.9		13.80
20	14.2	16.4	18.7	17.4	11.1	17.18
30	20.0	20.5	23.4	21.8	14.6	21.32
50	31.5	27.6	31.3	29.5	20.7	28.48
70	36.5	33.9	38.2	36.3	26.2	34.87
100	46.9		48.1	45.6	33.7	43.70
200	70.0	68.6	77.3	74.1	56.3	70.15
400	123.0	115.9	131.0	124.4	97.6	118.13

Table 2. Comparison of percentage errors relative to experimental data for Au ions implanted into SiO_2.

	Our study	PRAL	WS	SRIM	LSS
Maximum error (%)	21.00	15.49	31.69	22.53	34.88
Average error (%)	6.86	7.10	11.68	7.27	25.62
Minimum error (%)	0.22	2.00	0.63	0.54	19.57

reasonable results within considered energy range for Au ions implanted into SiO_2 when compared with experiment.

Figure 2 shows a comparison of our results for Bi ions implanted at the energies for incident ion from 10 to 400 keV into amorphous SiO_2 with SRIM, LSS, WS and with the experimental data taken from [10]. Although our results match the SRIM results for energies above 50 keV, for the lower energies our results are in better agreement with the experiment. Figure 3 compares the experimental values of the projected ranges of Br ions [20] with the calculation results for implantation energies from 50 to 400 keV. Agreement is obtained between the experimental and the calculated projected range. SRIM results seem in better

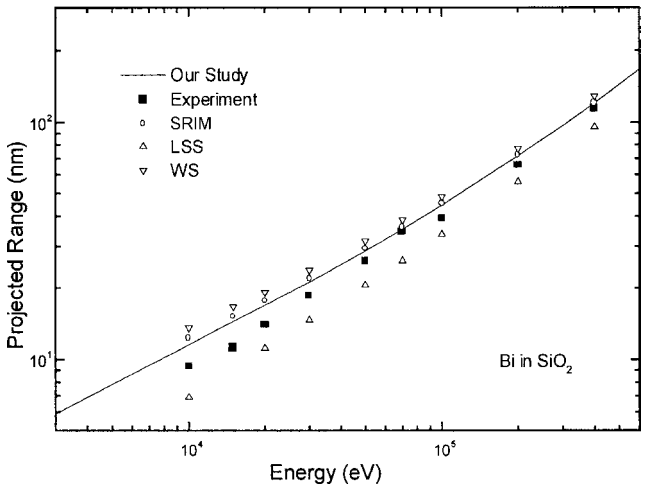

Fig. 2. Comparison of calculated values of the projected range $\bar{R}_p$ with SRIM, LSS, WS with experimental data of Bi ions implanted into SiO_2 at energies between 10 keV and 400 keV. The solid line represents the results of this study; the squares represent the experimental data of [10] and the meaning of other symbols were given in the Figure.

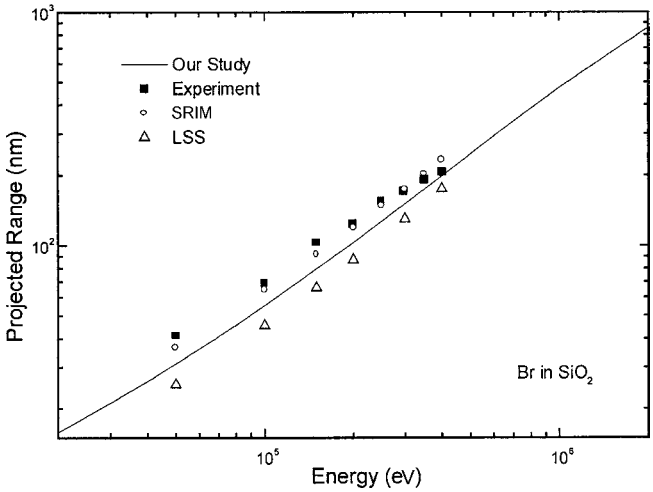

Fig. 3. Comparison of experimental, SRIM, LSS and calculated values of the projected range $\bar{R}_p$ of Br ions implanted into SiO_2 at energies between 50 keV and 400 keV. The solid line represents the results of this study; the squares represent the experimental data of [20]. Circle and up triangle represents SRIM and LSS, respectively.

agreement with experiment for the case of Br ions implanted into SiO_2. However, our results are situated between LSS predictions and SRIM for this case. Table 3 lists the comparisons between the experimental data, Monte Carlo simulation SRIM [22], LSS procedure, WS and our results for Bi ions implanted at energies from 10 to 400 keV.

An inspection of the average errors for Bi projectiles shows that the average error between our results and the experiment is 12% whereas the results of WS differ from experiment by about 27%. A comparison of the maximum errors reveals the fact that our results are again better than those of WS, as being 21% and 44% for the latter. In the case of the minimum error, our calculation and WS gives 5% and 12%, respectively.

Finally, the calculated values for mercury ions implanted into SiO_2 are shown along with SRIM, LSS and with experimental data of [18] in Fig. 4. An inspection of Fig. 4 clearly indicates that perfect matching exists between the

Table 3. Comparisons between experimental data, Monte Carlo simulation SRIM, LSS procedure, WS and our results for Bi ions implanted into SiO_2.

Energy (keV)	Experiment (nm)	SRIM (nm)	LSS (nm)	WS (nm)	Our study (nm)
10	9.4	12.3	6.9	13.5	11.4
15	11.2	15.1		16.6	
20	14.0	17.6	11.1	19.1	16.8
30	18.5	21.9	14.5	23.8	21.2
50	26.0	29.6	20.6	31.6	28.7
70	34.5	36.1	26.1	38.6	35.4
100	39.5	45.2	33.6	48.3	44.6
200	65.5	72.7	55.7	77.0	71.8
400	115.0	121.3	95.8	129.0	120.8

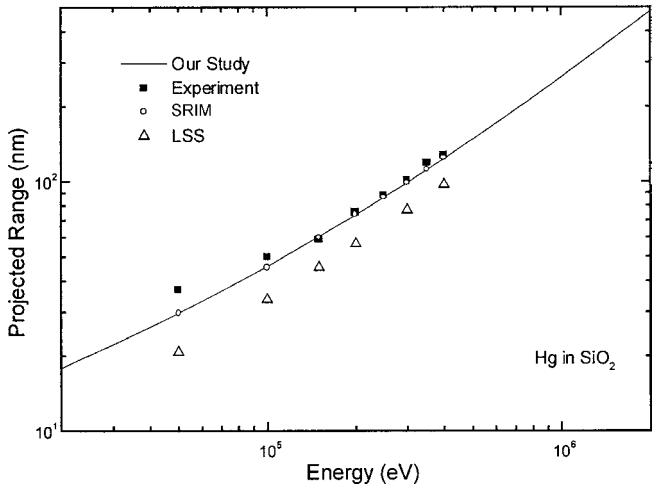

Fig. 4. Comparison of calculated values of the projected range of mercury ions implanted into SiO_2 as a function of ion energy.

calculated and the experimental values for the energies above 200 keV and general agreement was found between SRIM and our results.

Conclusion

In this study, a simplified method based on KRAL equation has been developed for the easy and efficient calculation of the projected range in diatomic targets such as SiO_2. This procedure can be used for prediction of ion implantation profiles. Although, Monte Carlo programs calculate ion ranges and angular distributions quite well, the major disadvantage of this method is that it is inherently a computer time-consuming procedure since a large number of ions is required to simulate only one energy. Our method is simpler and in some cases also more accurate than other comparable methods. In this method, running a single program range of ions for various ion target combinations and a wide range of energies can easily be found in a short time. This method uses nuclear stopping power presented by Ziegler *et al.* [23], electronic stopping power equation found by Montenegro *et al.* [13], a Runge–Kutta ODE solver of higher order. The calculated values of the projected range of some heavy ions in SiO_2 have been compared with the experimental data and other techniques such as Monte Carlo program SRIM, PRAL, LSS and WS. The comparison shows that the calculated results are in close agreement with experimental data and commonly used procedures such as SRIM and in most cases shows an average error around 10%. The present method has shown to be easy to handle and in some cases is more accurate than other comparable methods and it can be used to reliably predict heavy ion ranges in compounds such as SiO_2.

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