6.1 SCIENTIFIC MOTIVATION AND PROBLEM FORMULATION

In the recent days, energetic ions prove to be a powerful and versatile tool to modify and characterize the materials through Ion Beam based Analytical (IBA) techniques viz. RBS, NRA, ERDA etc [1-4]. The energy loss profile of energetic ions obtained through these techniques is simulated mainly via SIMNRA [5], RUMP [6] and NDF [7] computer codes. These codes are developed with the approximation that energy loss profiles are always Gaussian in shape and described in terms of average energy loss (first order moments) and energy loss straggling (second order moment). However, some recent experimental studies [8-10] show that the energy loss profile are not always Gaussian in shape but sometimes appear non-Gaussian also. Further, it reveals that the shape (Gaussian/non-Gaussian) of energy loss profile depends upon the thickness of the target material or fractional energy loss. For moderate thickness of the target material, the energy loss profile is Gaussian in shape while in case of very thin and very thick target materials, energy loss profile shows a non-Gaussian shape [11-15]. In order to analyze such spectra (Gaussian/non-Gaussian) more precisely, in addition to energy loss and energy loss straggling, two other higher order moments (Skewness and Kurtosis) should also be considered [8-10, 16-25], which are generally ignored. The study of these associated moments along with the energy loss of energetic ions is highly essential in order to improve the quantitative analysis of the computer simulation codes. Further, such study is also important to enhance the basic understanding of ion-solid interaction processes.

As far as energy loss is concerned, numbers of research groups are involved to study these moments both theoretically/experimentally for different ion-target combinations in various energy domains [26-48]. However, for associated higher order moments (Energy loss straggling, Skewness and Kurtosis), the status is quite different as compared to energy loss. The available experimental data points of energy loss straggling are not only haphazard but also fragmentary in nature. Theoretically also, the measured limited energy loss
straggling data points are not well proved with the available energy loss straggling theories, particularly in case of heavy ions. This is due to the reason that most of the available theories [38, 49-53] exclude the charge-exchange energy loss straggling component by assuming the incident ion as a fully stripped ion. Further, charge-exchange processes of incident ions are also a dominating part of the total energy loss straggling, so this effect should also be included theoretically [54-55]. Therefore, experimentally, a systematic measurements of energy loss straggling and theoretically, incorporation of charge exchange component with collisional component is highly essential. Further literature reveals that related to skewness and kurtosis, very few research publications are available [8-10, 16-22].

In addition to this, experimentally, it is almost impossible to cover entire range of ion-target combinations at various energies because number of such combination is infinite. So, only alternate is to use theoretical/semi-empirical formulations to predict these parameters. Further, before using these parameters, as input in computer codes, we have to authenticate the predicted values through comparison with the measured values and identify/develop the most suitable theoretical/semi-empirical formulation for such predictions.

The above discussion motivates us to construct a research problem based on the ion-matter interactions to unveil the mystery of passage of energetic ions through the target medium, particularly for those ion-energy combinations which are still uncovered. In the present thesis, our aim is to study energy loss and higher order moments (energy loss straggling, skewness, kurtosis) in energy loss distribution of energetic ions in different metallic foils.

6.2 HIGHER ORDER MOMENTS IN ENERGY LOSS DISTRIBUTION: A CONCEPTUAL FRAMEWORK

When energetic ions pass through the given thickness of the target materials, they loss their energies through various interaction processes, which depend upon the nature of incident ions (atomic number, energy) and target (atomic number, density) material. The most dominating interaction process,
through which the ions mainly lose their energies, is known as electronic energy loss process. In this process, energetic ions interact with the electrons of the target atoms and transfer their energies to eject or excite the target electrons from their orbits. Further, the electronic energy loss is thickness dependent so it is more appropriate to quote average energy loss per unit path length (*first order moment*) in place of average energy loss only. Also, the electronic energy loss process is energy dependent and most prominent at energy above 1MeV/n.

Due to statistical nature of ion-matter interaction processes, the energy loss fluctuates around the average energy loss, which is conventionally known as energy loss straggling (*second order moment*). Further, the energy loss straggling is divided into two components depending upon the nature of incident ions i.e. either the ions are fully stripped or partially stripped. For fully stripped heavy ions, the fluctuation (collisional energy loss straggling) arises due to the statistical variation in both the number of collisions with the target electrons and the energy transfer to each electron. In addition to these fluctuations, for partially stripped heavy ions, the fluctuation also arises as a result of variation of the ionic charge states, due to electron-capture and electron-loss processes, which lead to the so-called charge-exchange energy loss straggling. Both these straggling components are treated independently for theoretical calculations and finally to be summed in quadrature.

Both these moments (*energy loss and energy loss straggling*) are sufficient to understand the ion-matter interaction, for Gaussian shape energy loss distribution. However, for non-Gaussian distribution, higher moments are also needed. These higher moments are *Skewness* and *Kurtosis*.

*Skewness* (third order moment) comes into picture when energetic ions pass through very thin (where fractional energy loss, $\Delta E/E < 10\%$) and very thick (where fractional energy loss, $\Delta E/E \sim 50 - 90\%$) target materials. For very thin target material, the energy distribution spectra tails towards higher energy side (positive skewness) and for thick target spectra tails towards lower energy side (negative skewness) [11-15].
Fourth order moment, Kurtosis, measures the extent to which shape of energy distribution is more peaked (leptokurtic) or more flattened (platykurtic) than the Gaussian curve. For Gaussian energy distribution curve, the value of kurtosis is 3. Higher value indicates the leptokurtic curve and the value less than 3 indicates that curve is platykurtic.

6.3 CURRENT STATUS OF THE PROBLEM

The literature reveals that various theoretical and experimental research groups are involved in the study of energy loss of energetic ions in different target materials in different energy regions. On theoretical side, a variety of well known theoretical/semi-empirical formulations (LSS theory [39], Hubert et al. [40], Diwan et al. [41], Benton and Henke [42]), computer codes (SRIM [43], CasP [44], PASS [45-46], MSTAR [47]) and data tables (Northcliffe and Schilling [48]) are available. On the other end (experimental side), experimentalists generated the energy loss data for different ion-target combinations by utilizing different experimental techniques and accelerator facilities [26-37]. Further, through various studies, the available measured energy loss values are compared with the prediction of theoretical/semi-empirical/ computer-codes/data-table and merits and demerits of these formulations are highlighted. In order to use the prediction of these theoretical formulations in authenticated way, still, more experimentally measured energy loss values are required, particularly for those ion-energy-target combinations, where no experimental data is available. This newly generated data will be helpful to identify the best energy loss formulation and to refine the existing formulation.

As far as energy loss straggling is concerned (second order moment), the status is quite different from energy loss (first order moment). Experimentally, very limited energy loss straggling values are available, which are also fragmentary in nature. In order to simplify the mathematical treatment, energy loss straggling, is divided mainly into two components (collisional and charge-exchange) and treated independently. Conceptually, collisional straggling arises
due to the fluctuations of number of collisions and the amount of energy transferred in each collision, while charge exchange straggling originates due to variation of ionic charge states due to electron capture and electron loss processes. Till now, most of the developed energy loss straggling theories (Bohr [38], Lindhardh & Scharrff [49], Bethe-Livingston [50], Titeica [51] and Chu [52-53]) predict only the collisional component of the energy loss straggling. In addition, adopting the concept of effective charge in Chu theory [52-53] and adding the contribution of correlation effect, Yang et al. [54] developed empirical formulae for total energy loss straggling. It is observed, from the literature, that the deviation of the predicted values from measured values increases with increase of ion atomic number (Z₁) and thickness of the target material. This is due to the reason that, in this situation, in addition to collisional component charge-exchange component also plays a significant role in the total energy loss straggling. Therefore, there is a great need to perform systematic measurements of energy loss straggling for different ion-target combinations with the objective to check/refine the collisional straggling components and generate semi-empirical formula for charge-exchange component.

The studies of next higher order moments (skewness and kurtosis), which complete the story behind the ion energy loss process is too scarce. Till now, very few research papers are published related to the skewness measurements associated with the energy loss profile of energetic ions [8-10]. In literature, only single theoretical relation based on Bohr’s theory [38] is available for the prediction of skewness and kurtosis. Therefore, it is highly essential to measure skewness and kurtosis precisely and develop the theoretical/semi-empirical relations.

6.4 OBJECTIVES

The specific objectives of the thesis are:

1. To measure the energy loss and range of energetic ions with Z₁ = 2 - 8 in different metallic foils covering Z₂ = 13 - 79, in the energy region ~ 0.25 – 7.0 MeV/n.
2. To identify the best energy loss and range formulation through comparison between measured and predicted values, adopting most commonly used formulations viz. Benton and Henke, Northcliffe and Schilling, Hubert et al., Diwan et al., Paul and Schinner (MSTAR Code), Grande and Schiwietz (CasP code), Ziegler et al. (SRIM Code), Sigmund and Schinner (ICRU report 73).

3. To measure the energy loss straggling of energetic ions with $Z_1 = 2 - 8$ in varying thicknesses of different metallic foils.

4. To recognize the best energy loss straggling theory by comparing measured values with the computed values based on different straggling theories developed by Bohr, Lindhard and Scharff, Bethe and Livingston, Yang et al. and Titeica.

5. To develop analytical expression for charge-exchange straggling.

6. To see the dependence of energy loss straggling on energy loss.

7. To measure higher order moments (skewness and kurtosis) in energy loss distribution and compare with the prediction of Bohr’s theory.

In order to accomplish the above said objectives, we have considered He, Li, C and O ions in varying thicknesses of Al, Ti, Ni, Ag, Tb, Ta and Au metallic foils. These experiments are performed by utilizing 15 UD Pelletron accelerator facility at Inter University Accelerator Centre (IUAC), New Delhi, India.

6.5 MAJOR OUTCOMES OF THE PRESENT STUDY

The outcomes of the present study are described as under:

1. Huber et al. formulation along with its extended version (Diwan et al. formulation) provides excellent (within 5%) agreement with the measured energy loss ($-dE/dx$) and range values for Li, C and O ions in Al, Ti, Ni, Ag, Tb, Ta and Au metals.

2. Ziegler et al. (SRIM-2013.00 code) formulation shows better (within 8%) agreement with the measured values as compared to other considered energy loss ($-dE/dx$) and range formulations for all the ion-metal combinations, considered in the present study.
3. For the presently considered ion-metal combinations, measured and computed energy loss values do not show $Z_2$-oscillations effect in energy loss ($-dE/dx$), in the energy region $\sim 0.50 - 7.00$ MeV/n.

4. The comparison between measured and predicted energy loss straggling values show that the results of Titeica collisional theory are in better agreement with the measured values as compared to other collisional theories for He (1.37 MeV/n), Li (5.68 & 7.11 MeV/n), C (4.51 & 7.01 MeV/n) and O (4.91 & 6.16 MeV/n) ions in Al, Ti, Ni, Ag, Tb, Ta and Au metallic foils, in the fractional energy loss ($\Delta E/E$) limits $\sim 5 - 95\%$.

5. An analytical fitted expression for charge-exchange component of energy loss straggling as a function of effective charge of the incident ion ($Z_1^*$), fractional energy loss ($\Delta E/E$), incident energy of the ion ($E$) and atomic number of the target material ($Z_2$) is developed.

6. Theoretical results based on the presently developed analytical expression (charge-exchange component) in combination with Titeica theory (collisional component) provides reasonable agreement within $\sim 20\%$ with the measured energy loss straggling values for all the considered ion-metal combinations, in the fractional energy loss ($\Delta E/E$) limits $\sim 5 - 95\%$.

7. The linear relationship between energy loss straggling and energy loss is observed irrespective of the incident ion, its energy and metal thickness.

8. The measured Skewness results reveal that the considered energy loss distributions are slightly positively skewed for very low fractional energy loss and slightly negatively skewed for very high fractional energy loss ($\Delta E/E$) limits. However, for moderate fractional energy loss, energy loss distributions are almost non-skewed (symmetrical).

9. Kurtosis measured values indicate that the shapes of considered energy loss distributions are about mesokurtic type.

10. The behavior of measured higher order moments (skewness and kurtosis) follows the predictions of Bohr’s theory.

These outcomes are important from both the fundamental as well as applied perspectives. From fundamental perspectives, the basic knowledge of
these moments is important: (a) for complete understanding of the profile of energy loss distribution, (b) for the refinement of the existing theoretical formulations and (c) to develop new formulation. Further, these outcomes are utmost important as an input for computer simulation programs, which are exclusively used for data analysis of ion beam analysis methods.

6.6 FUTURE PROJECTIONS

Some of the future projections of our work are:

1. To extend the measurements of energy loss and associated higher order moments for incident ions covering $Z_1 = 9 - 29$ in varying thicknesses of different metallic foils.

2. To develop the expression for evaluation of effective charge as a function of incident ion parameters without resorting any empirical parameterization.

3. To extend an analytical expression, for charge exchange straggling which will be applicable for both elemental and complex polymeric materials.

4. To verify, whether the shoulder effect predicted theoretically by Sigmund [23-24] in energy loss straggling versus energy curve is reproduced experimentally or not.

5. To develop a suitable model to predict the skewness and kurtosis associated in the energy loss distributions of energetic ions.

6. To study how the energy loss and energy loss straggling phenomena in case of cluster ions are different than that of heavy ions.
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