PART I

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1. Scope and object of the work

In the binary fission process, the fissioning nucleus splits into two excited fragment nuclei. Formation of compound nucleus, its division into fragments as a result of the re-arrangement of the nucleons, the formation of fragments with varying nuclear characteristics are the sequence of events in the fission phenomenon. One of the most characteristic features of low energy fission in the actinide region is the predominance of asymmetric division of the fissioning nucleus. Till very recently satisfactory theoretical explanation of this phenomenon was not forthcoming. The present work is undertaken to study this characteristic of the fission process using the Order-Disorder Model. Physical characteristics of the fission product species formed in fission has several possible practical applications. Experimental study of some of these characteristics, specially at short times after fission has also been carried out and their application to fissile material identification and estimation techniques has been explored.

2. Review of the work done by earlier workers

A critical review of work done by earlier workers on the topics dealt with in the thesis is given below.
2.1 Theories and models on fission process

First theoretical studies of the fission process were based on the Liquid Drop Model (LDM) and were given by Bohr and Wheeler (2) and Frankel (3). Though it had success in explaining certain aspects of the fission process like correlation of spontaneous fission half life with $A^2/A$ parameter and fission barrier calculation etc, it did not explain the striking and basic feature of fission, namely the asymmetry of mass division. The LDM continued to be the most accepted one with large number of semi empirical and sometimes arbitrary corrections and modifications.

Bohr (4) has observed that the life time of a compound nucleus is about a million times longer than the fundamental nuclear periods before it decays by emission of radiations or by fission. Fission takes place if there is in the process a sufficient concentration of potential energy due to deformation which enables the nucleus to pass over the saddle point, at which the repulsive coulomb force is just balanced by the cohesive nuclear interaction. If the excitation energy is not too far above the fission threshold, the nucleus is cold while passing over the saddle point, as major part of the energy content is bound in potential energy or deformation. These nuclides have an elongated shape even in their ground states and the ground state excitation may therefore resemble that at the saddle point.
In the statistical theory of Fong, fission process is slow enough so that nucleons might cross the nucleus several times from saddle point to scission. And hence it was assumed that statistical equilibrium is established at every instant or step of the process. Relative probability of a fission mode is then proportional to the density of quantum states of the nuclear configuration just before scission. This is calculated from excitation energy obtained from a mass formula and assuming $P_3$ type deformation (liquid drop model) for the impending fragments. Internal excitation energy as a function of fission mode showed a peak in the asymmetric fission region and this was attributed to $Z = 50$ and $N = 82$ shell closures. The peak in this function is said to cause asymmetry in fission. Charge distribution function for a given mass was derived to be approximately Gaussian with $\sigma = 0.51$. When Perrine and Storey applied Fong's prescription to $^{239}$Pu $(n_{th}, f)$ the predicted distribution was in poor agreement with observed experimental data. They tried to modify the mass formula used by Fong but the agreement became worse. Later Fong secured better fits by revising the parameters in his mass equation.

Wing and Fong calculated charge distribution in fission on the basis of the statistical theory using a nuclidic formula developed by them. They found that width of the charge
distributions \((s = 0.51)\) does not change with the new mass formula. According to this theory \(Z_p^{(m)}\) (most probable charge) for a mass is found by maximizing the excitation energy. It has been tacitly assumed in the formulation that \(Z_p\) function is smooth. But there are reasons to expect fine structures\(^{(1)}\) in this function as in the kinetic energy and \(\gamma(A)\) functions.

Newson\(^{(9)}\) calculated mass yields for a number of fission reactions using a similar approach as Pong. He assumed as a first approximation that probabilities are proportional to the product of level densities of the pair of fragments. A free parameter was used for the correction of excitation energies of fragments and for possible departure from equilibrium assumption of Pong.

Paissener and Wildermuth\(^{(10)}\) proposed a model of fission assuming that certain cluster formations take place in the compound nucleus before scission which remain essentially unaffected through the rest of fission process. Neutron and proton shells were considered for the specification of cluster structures. The cluster due to 82 neutron and 50 proton shells was considered for higher fragment peak. For the lighter peak it was ambiguous, and depending on the fissioning nucleus, either 50 or 40 neutron or 40 proton shells were expected to
play a role. A Gaussian curve was fitted to the experimental mass yield points. The final mass yield was then obtained by addition of such Gaussians from different cluster configurations. The distribution obtained was trapezoidal with a flat top and yield values at symmetric fission were vanishingly small, contrary to the experimental observation.

Norenberg\(^{11}\) used a molecular model to calculate the charge distribution in low energy fission. The compound nucleus is described as two interacting fragments separated by a distance large enough to treat the compound nucleus as a bi-nuclear molecule bound by other nucleons. The CBS wave function determined from the condition of minimum for total energy was solved. Solutions for various distances were arrived at. The mean charge distribution is presented for an effective fragment distance given by observed mean kinetic energy, assuming it to be given by coulomb repulsion. Wahl plot shows a minimum narrow band at \(A = 132\).

Ramanna et al\(^{12}\) treated fission as a Markov process in which a random transfer of nucleons takes place between the two sides (poles) of the fissioning nucleus from saddle point to scission. The final mass distribution is obtained assuming the configurations having magic number \(N = 82, Z = 50\) are particularly
stable. An equilibrium value is obtained with 500 steps of nucleon transfer. The staying in probability for the shell configuration are calculated by assuming a Gaussian for $Z$ and $N$ distributions about the shell. Ground state conditions are applied since a fissioning nucleus is essentially cold at scission. It is observed that near the saddle point both protons and neutrons take part in the random process but after a short time the protons get polarised to the ends of the fissioning nuclei leaving only neutrons to take part in the random walk.

Ramamurthy and Ramanna\(^{(13)}\) calculated the mass and charge distribution by approximating the fissioning nucleus as two nearly independent entity in close proximity. The continuous rearrangement of the nucleons are slow so that the process is treated as a time dependent stochastic process up to the scission point, thermodynamic equilibrium and the condition of minimum potential energy are assumed. There is qualitative agreement with experimental results.

By using 'shell' correction term with LDM potential, Strutinsky\(^{(14)}\) has shown double humped barrier in the variation of nuclear potential energy with deformation, in which the second minima predicted the existence of spontaneously fissioning isomers, which has been found in experiments by other workers\(^{(15,16)}\).
Strutinsky's prescription of shell correction with deformation has been used by Ignatyuk\textsuperscript{(17)} to predict mass distribution in fission. The distribution was too narrow compared to the observed yield distribution and the peak position was six units lower in mass value.

Denschlag and Qaim\textsuperscript{(18)} proposed a dumb bell shape model for the transition state configuration in low energy fission. They assumed that there are two spherical configurations of 50 protons and 82 neutrons in the heavier side and 50 neutrons and 32 protons in the lighter side leaving 10 protons and 12 neutrons in the neck. Fission reaction yielding fragment masses 132 to 154 and 82 to 104 exclusively takes place in the neck. The point at which scission takes place within the neck determines the mass distribution. Mass yield and distributions are correlated in the range of $A = 132$ to 154 to the number of nucleons ($d$) from the neck with the spherical part. The inflexion point of increasing mass yield coincides with $d = 0$. The $\nabla$ is minimum for $d = 0$ i.e. when the spherical part alone goes as one fragment. $Z_p$ for $A = 132$ is taken as 50 ($Z_p - Z_{UOD} = 1.5$) and that for $A = 82$ is taken as 32 ($Z_p - Z_{UOD} = 0$). This resulted in a straight line which is claimed to have a similar slope as the experimental "Wahl plot".
Neither the fine structures nor the behaviour of the Wahl plot in other mass regions are attempted to be explained.

The order-disorder model (ODM)\(^{(1,19)}\) proposed by Iyer and Ganguly envisages that the fissioning nucleus undergoes charge polarization into two impending fragments each with number of neutrons corresponding to the ground state beta-stable nuclides. The balance neutrons are shared between the two polarised impending fragments as in order-disorder process\(^{(20,21)}\). By applying thermodynamic concepts to this system and deriving the condition for free energy minimum, isotopic distribution (i.e. probability distribution of fragments having same charge number with different neutron numbers) for each charge was worked out. These distributions together with the experimental fragment mass yields give the independent yields. It has been used to work out a wide range of fission data such as total isotopic yield distribution, total isotopic yield distribution, charge distribution parameters etc. Correlation including odd even structures in the total isotopic yield with that observed was good.

The isotopic probability distributions of the fragments as obtained in the above work along with similar distributions for products calculated from product charge distribution parameters; a new procedure was developed by Iyer and Ganguly\(^{(22)}\) for
obtaining the number of neutrons $\bar{\nu}(z,A)$ evaporated from individual fragments. This parameter worked out for the first time shows shell-effect in neutron evaporation process e.g. at $Z=50$ and $N=82$, $\bar{\nu}(A), \bar{\nu}(Z), \bar{\nu}(N)$ distributions calculated therefrom were also presented. The kinetic energy distributions for individual fission fragments and their isobaric, isotopic and isotonic totals were also presented. Average fission parameters and the fission neutron spectrum show good agreement with experimental data.

A computational procedure based on the ODM for predicting independent yields of fission products in fission of $^{235}U$ induced by high energy neutrons has been described by Sharma et al. The computational procedure involves a scheme for distribution of extra excitation energy originating from the kinetic energy of the neutron absorbed and its binding energy to the fissioning nucleus between the two impending fragments. Excitation energy of the fragments for high energy neutron induced fission is obtained by adding the extra excitation energy to the base values for the spontaneous fission case. The $\bar{\nu}(z,N)$ obtained from this, using the evaporation scheme along with the fragment isotopic distributions given by ODM gives the product isotopic distributions. Independent yields of product are then calculated from these distributions.
and mass yield data. A number of parameters like total isotopic yield distribution, charge distribution parameters \( \nu(z), \nu(A), \nu(N) \) are computed for 2 MeV and 14 MeV fission of \(^{235}\)U. The effect of magic numbers on \( \nu(z,N) \) is found to be less than that is found in case of thermal fission.

Maruhn et al\(^{(24)}\) introduced a two centre shell-model of fission describing nuclear shapes occurring during the fission process. In the calculations quadrupole deformations and mass asymmetric distortions are taken as parameters. Potential energy surfaces are calculated using the macroscopic-microscopic method prescribed by Strutinsky. The parameters are taken as dynamic collective coordinates and the model is used for generating mass parameters in the cranking model approximation. Schrödinger equation for movement in the mass-asymmetry degree of freedom is solved numerically, which is then interpreted as the main component determining the fragment mass yield in spontaneous fission. It was concluded that potential energy is the main factor determining mass yield and the dynamic treatment would have only second order effects. The results obtained using \( \lambda \) (overall length of the nucleus in units of diameter of the corresponding spherical nucleus) equal to 1.65 gives better agreement with the experimental data for \(^{236}\)U than that obtained
yields in
with $\lambda = 1.8$. In both the cases however the symmetric region are about four orders of magnitude lower than the expected value.

Mustafa et al. (25) have proposed a two centre shell model for fission to explain asymmetric deformation in fission. Four independent shape variables are used for potential energy calculations. These calculations are carried out for $^{202}$Pb, $^{210}$Po, $^{236}$U, $^{248}$Cm, $^{252}$Fm, $^{258}$Fm and $^{264}$Fm. Asymmetric fission is found to be preferred energetically for $^{236}$U, $^{248}$Cm and $^{252}$Fm and symmetric fissions are preferred in $^{202}$Pb, $^{210}$Po, $^{258}$Fm and $^{264}$Fm. However fission for $^{202}$Pb and $^{210}$Po corresponds to high energy fission whereas latter ones are for spontaneous fission and as such are not strictly comparable cases. The preference of symmetric mass division for $^{264}$Fm is very strong due to double magic effect (Z=50, N=82) formed at the symmetric mode. In general the structures in the potential energy surface are due to an interplay between compound-nucleus shell-structure, fragment shell structures and liquid-drop model energies. It has been stated that comparison with experimental results indicated that mass distribution can be correlated with the potential energy surface in the neighbourhood of scission.

Jensen and Dossing (26) presented a statistical calculation for mass distribution of fragments as a function of excitation.
energy from saddle point properties of fissioning nucleus. The quantities involved in the formulations are the potential energy surface, level density as a function of deformation, angular momentum and excitation energy. Two symmetric deformation parameters and one left-right asymmetry parameter are taken to span the deformation space. The mass distribution of the fission fragments is assumed to be determined around the second saddle point and given by statistical arguments. In case of $^{240}$Pu it is shown that, as one goes away from the saddle point, the correlation between observed and calculated distributions disappears. No fine structures are seen in the distribution of $^{240}$Pu. The distributions for $^{258}$Fm is identified to be asymmetric whereas that for $^{226}$Ra has been predominantly symmetric. The width of the peak for low energy fission is around a factor of two too narrow compared to the observed data.

Pauli and Ledergerber\textsuperscript{(27)} discussed a model formulation for the dynamics of fission. The model is checked by calculating spontaneous fission life time and claimed to have reasonable agreement with experiments. It is stated that fission barriers are dynamic and preformation of the nascent fragments arises in the parent nucleus around the outer barrier. Discrepancies of the various approaches are indicated and are attributed to the
utter complexity of the nuclear many body problem "which has not left much choice for doing better and that rules are desperately needed not to get lost in the flood of data".

Fong\textsuperscript{(28)} has investigated the problem of asymmetric mass distribution in fission using a dynamical theory under a variety of conditions which covers most of the conceivable cases. It has been observed that the results obtained by the various ways cannot explain the experimental results of asymmetric mass distribution and the difficulties are intrinsic to the nature of the theory and are not likely to improve only by improvement of mathematical treatment. This apprehension is in agreement with the observation of Maruhn et al\textsuperscript{(24)} (cf. P.10).

Recently Wilkins\textsuperscript{(29)} et al proposed a model of nuclear fission based on the statistical equilibrium among the collective degrees of freedom at scission point. Relative formation probabilities of the fragment pairs are determined from the relative potential energies of a system of two nearly touching coaxial spheroids with quadrupole deformations. The total potential energy of the system is calculated as the sum of liquid-drop, shell and pairing correction terms for each spheroid and coulomb and nuclear potential terms describing the interaction between them. A single choice of
values for the distance between the tips of the spheroids, intrinsic excitation energy and the collective temperature of the fragments are made in the calculation. The mass distributions worked for various fissioning nuclides show asymmetric distributions and bunching of higher mass peaks. However the widths of the peaks are too narrow and the peak-to-valley ratios are much less (e.g., about twenty-five times lower in case of $^{235}$U) than the observed values. Charge distributions and kinetic energy distributions of the fragments are also worked out. Lack of agreement with experimental data is attributed to the expected errors of shell-corrections obtained using Strutinsky's prescription.

Hyde$^{30}$ speculated fission as a rate process as envisaged by Swiatecki and the rate equation for fission is expressed as

$$ R = A e^{-E/kT} $$

where $E$ is the fission threshold energy, $A$ is the frequency factor and $kT$ is the nuclear temperature. The equation is analogous to that in the rate process theory of chemical reaction. The analogy drawn for this is as follows:

<table>
<thead>
<tr>
<th>Chemical Reaction</th>
<th>Fission</th>
</tr>
</thead>
<tbody>
<tr>
<td>Activation energy</td>
<td>threshold energy</td>
</tr>
<tr>
<td>Reaction rate</td>
<td>fission width</td>
</tr>
<tr>
<td>Adiabatic hypothesis</td>
<td>disregard of internal degrees of freedom</td>
</tr>
</tbody>
</table>
But he has not reported the use of the expression for detailed calculations of any fission parameter.

Third et al.\(^{(31)}\) used Myrings's absolute reaction rate theory of chemical reaction for calculating relative independent yields of two isobars using an expression of the form

\[ e^{-\Delta M/\theta} \]

where \(\Delta M\) is obtained from the difference of fission energy of two modes and \(\theta\) is the nuclear temperature. It is stated that excitation energies for such divisions are same and hence the entropy difference is zero, which may not be true. The temperature is taken as a free parameter and is equal to 1.4 MeV. This is an arbitrary stipulation.

Norenberg\(^{(32)}\) discussed various specific models on fission to have a unified approach on the theories. Static scission point model, statistical model, adiabatic model, di-adiabatic model and thermodynamic or semi-equilibrium model, are discussed. In the semi-equilibrium model, the probability of element distribution was given by

\[ P(n)_{z_1, z_2} \propto \exp \left\{ -\frac{1}{\beta_{\text{coll}}} \varepsilon_{z_1, z_2}^n \right\} \]

for the fission band \(n\), where \(\varepsilon_{z_1, z_2}^n\) is the adiabatic energy.
at scission point and 
\[ \frac{k_B}{\beta_{\text{cal}}^n} = \sqrt[n]{T_{\text{cal}}^n} \]  
\[ k_B \] is the Boltzmann constant. No specific computational results using the formulation was presented for the elemental distribution.

2.2 Mass and charge distribution in fission

Mass distribution of fission products is measured radiochemically and a vast amount of data are available (33-39). Von Gunten (40) has studied the general behaviour of mass distributions of various fissile elements for spontaneous as well as for neutron induced fission. The observed common features and regularities of the distributions can be summarised as follows:

(i) distribution of mass-split is highly asymmetric in the region from actinium to californium; The asymmetry gradually decreases with higher atomic charge and mass numbers;

(ii) peak position of the heavy mass group remains more or less constant. It is found to shift slightly for \( A > 250 \);

(iii) peak positions in the light mass group shift towards the heavy mass group with increasing mass of the fissioning nuclide;

(iv) width of the valley reduces with higher mass and charge number of the fissioning nuclide but the width of the peak increases with increasing mass number of the fissile nuclide.
(v) peak to valley ratio reduces with increasing mass and charge number of nuclide;

(vi) peak to valley ratio also reduces with higher excitation energy of fissioning nucleus.

Though the error in the low yield symmetric region is large compared to high yield region, best results for symmetric and highly asymmetric regions are obtained using radiochemical method.

According to Wahl (37) 50 proton and 82 neutron shell edges are often associated with the constancy of the position of the heavier peak. Von Gunten (40) also comes to the same conclusion.

Asymmetry in the fission mass yield distribution is often expressed in terms of peak-to-valley ratio of the distribution. With increasing excitation energy of fissioning nucleus, the fine structures tend to disappear, the probability of symmetric yield increases and peaks are broadened (37). Peak-to-valley ratio of \(^{235}\text{U}(n_{th},f)\) is 630(36) as compared to the value of 7 for 14 keV neutron induced fission of \(^{235}\text{U}\).

Yields of primary fragments can be determined by physical methods either by correlation of kinetic energies (pulse heights from solid state detectors) or velocities (time of flight) of fragment
pairs. Typical of these are the double velocity measurements of Milton and Fraser (41) and correlated kinetic energy measurements of Schmitt et al (42). Unik et al (43) have also made fragment mass and kinetic energy determinations for seven thermal neutron induced fission reactions and six spontaneous fission reactions. The structures in the distributions are attributed to the enhanced formation of even \(Z\) fragments. The results obtained by ODM (4) pointed to the same conclusion.

Papas et al (44) reviewed the methods of fragment mass yield distributions. Instrumental methods do not attain peak to valley ratio of 600 for \(^{235}\text{U(n, f)}\) as obtained by radiochemical methods. The best reported value is about 450, and this is attributed to false events registered in the high slope and valley regions due to the limitation in mass resolution in the experimental technique.

Charge distribution is generally deduced from radiochemically determined independent yields of fission products. It is usually discussed in terms of yield distribution of isobaric fission products with charge. It has been attributed empirically to follow a Gaussian distribution:

\[
\left( \frac{c_A}{\sigma} \right)^2 \exp \left[ -\left( Z - Z_p \right)^2 / \sigma_A \right]
\]
where $C_A$ and $Z_p$ are obtained empirically. Several hypotheses for the charge distribution like, the postulate of unchanged charge density ($UCD$) and equal charge displacement ($ECD$) have been proposed but these do not fit the experimental data very well.

Wahl\(^{(33)}\) proposed the plotting of $Z_p - Z_{UCD}$ (Wahl plot) vs fragment mass for studying the most probable charge, where $Z_{UCD}$ is the charge for a mass given by the unchanged charge density. Following the proposal of Pappas\(^{(45)}\) that since charge distribution is decided in the fissioning nucleus it would be appropriate to express the charge distribution in terms of fragment mass.

Width of the Gaussian distribution for products is found to vary with mass. Due to paucity of data many authors have used constant width for the distributions\(^{(36)}\).

Some authors (for example Gordon and Aras\(^{(46)}\)) assume the width of the fragment charge distribution to be constant with mass and attribute the observed variation for products to neutron evaporation. No theoretical basis for the constancy of width exists. Wahl\(^{(34)}\) in fact does not recommend fitting of Gaussian distribution since this removes the structures in these distributions. Experimentally slight skewness in the
distributions is also observed. In the QDM(1) this skewness in the isotopic distributions arises naturally in the model. Further the variation in spread in the isotopic distribution is also natural in the model. These effects are then reflected in the isobaric charge distributions obtained therefrom.

Instrumental methods of investigation of charge distributions use a gas filled mass separator (e.g.: Armbruster et al(47)) or simultaneously measure kinetic energy of complementary fission fragments and their characteristic X-rays (e.g. Glendenin et al(48)). According to Pappas(44), the results obtained by instrumental methods show apparent and unexplained discrepancies with those of radiochemical values even in trend. Because of limitation of mass resolution one cannot study the fine structures in \( Z_p \) function using these methods.

2.3 Investigation of Fission Product Gamma Spectra and Delayed Neutron activity for Fissile Material Identification and Estimation

Baumun w et al(49) studied the gamma spectra of fission products from the thermal fission of \(^{235}\text{U}\) and \(^{239}\text{Pu}\) using a Ge(Li) detector. The samples were irradiated for two minutes and counted for 10 minutes following a decay of 30 minutes. The spectra cover energies from 0.6 to 2.2 MeV. Intensities of
certain peaks were found to have substantial differences. The peak at 1.248 MeV was six times higher in $^{235}\text{U}$ than in $^{239}\text{Pu}$. The peak at 0.767 MeV was twice as intense in $^{239}\text{Pu}$ as in $^{235}\text{U}$. The 0.724 MeV peak present in $^{239}\text{Pu}$ did not appear in $^{235}\text{U}$.

East and Keppin\(^{(50)}\) have reported detailed studies on the decay characteristics of fission product gammas and delayed neutrons and also spectral behaviour of fission product gammas. The fission product gamma spectra from $^{239}\text{Pu}$, $^{233}\text{U}$, $^{235}\text{U}$, $^{241}\text{Pu}$ were studied using a Ge(Li) detector after an irradiation of 5 minutes and decay time of 60 minutes. The measurements cover the energy range of 0.32 to 1.45 MeV. The 0.724 MeV line attributed to $^{105}\text{Ru}$ was found to be present in the $^{239}\text{Pu}$ gamma spectra but not in that of $^{235}\text{U}$. Certain other lines also showed different intensities for different cases. The decay behaviour of gross gammas and delayed neutrons for $^{235}\text{U}$ and $^{238}\text{U}$ were used to study the ratio of delayed neutrons and that of delayed gamma intensities for $^{235}\text{U}$ and $^{238}\text{U}$. These studies were however only for fast fission. Delayed gamma-to-delayed neutron ratios were not investigated.

Fisher and Angile\(^{(51)}\) used a pneumatic transfer system and a pulsed reactor for studying the time and energy dependence
of fission product gammas from the fast fission of a number of nuclides, in the time range of 0.2 to 45 seconds. They used a 4" x 4" NaI(Tl) detector. Using the response function of the detector they worked out the photon spectra. No significant changes in the spectra for the different fissile materials were noticed. The difference in structures, if any in the original spectra might have got masked by the transformation of the observed spectra to the photon spectra by dividing the energies into different bins.

Walton et al.(52) investigated the time dependence of gross fission product gammas from the photo fission of $^{238}$U and $^{232}$Th in the time range of microseconds to 32 milliseconds. However no spectral studies were made and no quantitative analysis of decay studies were reported.

3. Present Work

The concept of charge polarization in the fissioning nucleus as envisaged in the Order-Disorder Model proposed by Iyer and Ganguly(1) has been adopted in the present work to predict the asymmetric division of nuclear charge and mass for spontaneous fission of a number of nuclides. In this work an attempt has been made to predict various fission parameters.
without using any free or adjustable parameters or any fission data as input. The computational procedure uses stable neutron numbers of nuclei as input data. The work presented in the thesis is divided into three chapters.

In Chapter 1, the first step of the CDI, i.e., charge polarization in the early stages of fission process along with beta stable neutrons has been used to predict the relative probability of formation of fragments of different charges.

In the present formulation, fission is taken as a rate process as envisaged by fission for chemical reactions, having a frequency term and an energy dependent exponential term. A conditional stochastic process of polarization of $Z_L$ protons with $N_L$ neutrons and $Z_H$ protons with $N_H$ neutrons is taken into account for deriving the frequency factor. The general features of the probability distribution comes intrinsically from this frequency term. The exponential term introduces energy factor in the formulation of the rate process and brings about a good agreement with the observed data.

With the present formulation, total isotopic yields of various fissile elements have been worked out. The bunching of peaks around $Z = 56$ for various nuclides, the decrease of
asymmetry with increasing charge/mass of the fissioning nucleus are predicted in the present work.

In Chapter 2, the total isotopic yields obtained using the model in Chapter 1, together with the isotopic distributions (probabilities of fragments with same charge and different neutron numbers) computed using the second step of the QDM gives the independent yields of fragments. This when added along the mass line gives rise to mass distributions of the fission fragments. The results obtained for spontaneous fission of $^{236}_{\text{U}}, \, ^{240}_{\text{Pu}}, \, ^{246}_{\text{Cm}}, \, ^{252}_{\text{Cf}}, \, ^{256}_{\text{Fm}} \, \text{and} \, ^{260}_{\text{Au}}$ are presented and discussed. The total isotonic yields for $^{236}_{\text{U}}, \, ^{240}_{\text{Pu}} \, \text{and} \, ^{252}_{\text{Cf}}$ are also worked out. The charge distribution parameters, viz., the mean $Z_p$ and the width $C_A$ of the charge distributions are studied and are compared with experimental values.

In Chapter 3, studies of short-lived fission product gamma spectra and delayed neutron activities from the thermal fission of three common fissile materials, viz. $^{233}_{\text{U}}, \, ^{235}_{\text{U}} \, \text{and} \, ^{239}_{\text{Pu}}$ were studied to develop techniques for identification and measurements of these materials in a mixture. The techniques developed for the estimations have got potential for non-destructive assay application in fissile material management.

What follows in the rest of the thesis is the work carried out by the candidate.