STOCHASTIC ANALYSIS OF RANDOM GRAPH PROBLEMS.

Mathematicians who are not interested in graph for their own sake should view the theory of random graphs as a modest beginning to learn variety of techniques and try to prove more complicated random structure. The theory of random graph was founded early nineteenth century. BELA BOLLOBAS [9] has studied the extremal problems in graph theory using probabilistic methods.

In section 1, we present the evolution of random graphs with emphasis on the use of probabilistic method in extremal graph theory. We give an account of random graph problems in polymer chemistry in section 2. Whittle[60,61,62] has studied the polymerisation processes with intrapolymer bonding with several types of unit, two types of units and one type of units while examining the cosh model and poisson model. He has also given the connection between random graph and polymerisation processes[53,54]. We give an expository account of Whittle in section 3. There is a considerable nature of statistics of polymers induced by prescribed structure of bonding. We give the main results of different types of bonding and bond reactions in section 4. We also discuss the time dependent solution of the mode.

5.1 EVOLUTION OF RANDOM GRAPHS

Various Models of Random Graphs play an important roll in extremal graph theory. The two most common models are \( \mathcal{G}(n,M) \) and \( \mathcal{G}(n,P) \). Both consist of graphs on a fixed set \( V \) of \( n \) labelled vertices. \( \mathcal{G}(n,M) \) is the set of all graphs on \( V \) with
M edges, and all graphs are taken equiprobable. The underlying set of the space $\mathcal{G}(n, P)$ is $\mathcal{G}^n$, the set of all graphs on $V$, and the probability of a graph $G \in \mathcal{G}^n$ having $m$ edges is $p^m q^{n-m}$, where $N = \binom{n}{2}$ is the number of possible edges and $q = 1-p$. Thus the graphs in $\mathcal{G}(n, p)$ are constructed by choosing edges independently and with probability $p$.

Let us denote $G_m$ and $G_p$ for random elements of the sets $\mathcal{G}(n, M)$ and $\mathcal{G}(n, P)$; $P_m$ and $P_p$ denote the appropriate probabilities. Every graph invariant $x = x(G)$ becomes a random variable on our space of random graph and let us study the distribution of this random variable as $n \to \infty$.

**Definition 5.1.1** A subset $Q$ of $G^n$ is said to be a property of graphs of order $n$ if $G \in Q$, $H \in \mathcal{G}^n$, and $G \cong H$ imply that $H \in Q$. Given a model $\Omega_n$ of random graphs of order $n$, we say that almost every (a.e) graph in $\Omega_n$ has a certain property $Q$ if $p(Q) \to 1$ as $n \to \infty$.

**Definition 5.1.2** A property $Q$ is said to be monotone increasing, or simply increasing, if whenever $G \in Q$ and $G \subset H$ then also $H \in Q$. A property $Q$ is convex if $F \subset G \subset H$ and $F, H \in Q$ imply $G \in Q$.

Over the past few years a noticeable shift has been taking place in extremal graph theory towards probabilistic methods. The most obvious sign is that random graphs are used more and more, but that is not all. Even more significantly, a probabilistic frame of mind was needed to find many of the proofs, which on the surface have nothing to do with probabilistic ideas. In several beautiful and difficult proofs the underlying philosophy is that we do have to care about single vertices, say, for it.
suffices to make use of the fact that there are many subsets of vertices of a given

cardinality with the right properties. To give a simple example, one often makes use
of the fact that of $X_1, X_2, \ldots, X_N$ are nonnegative integers bounded by $A$,

$$
\sum_{i=1}^{N} X_i = Na \text{ and } 0 < b < a, \text{ then at least } (a-b)N/(A-b) \text{ of the } X_i \text{'s are greater}
$$

than $b$. Equivalently, if $X_i$ is a random variable, $0 < X < A$ and $E(X) = a$, then

(1) $p(X > b) \geq (a-b)/(A-b)$ for all $0 < b < a$. In equivalence (1) has the following reformula

tion in graph-theoretic terms. If $B$ is a bipartite graph with bipartition $(X,Y)$,

$X = \{x_1, x_2, \ldots, x_n\}, Y = \{y_1, y_2, \ldots, y_m\}, d(y_j) \leq \Delta \text{ for all } j, 1 \leq j \leq n,$

then for

$$
d' < d = \sum_{i=1}^{m} d(x_i)/n \text{ there are at least } (d-d')n/(\Delta - d') \text{ vertices } y_j \text{ of degree at least } d'.
$$

Needless to emphasize in the great majority of the cases the merit is in fending

the need for probablistic inequalities and apply them cleverly, and not in proving the
inequalities.

It is intuitively obvious, and happens to be true, that if $M$ or $p$ is increased then

the probability of a montone increasing property does not decrease. Further more, if

$pN,$ the expected number of edges in $G_p$ is close to $M$ then the models $\mathcal{G}(n,p)$ and

$\mathcal{G}(n,M)$ are practically interchangable.

**Theorem 5.1.1** (1) Let $Q$ be any property and suppose $pqN \rightarrow \infty$. Then the follow

ing two assertions are equivalent.

a) Almost every graph in $\mathcal{G}(n,p)$ has $Q$.

b) Given $x > 0$ and $\epsilon > 0$, if $n$ is sufficiently large then there are

$$
1 \geq (1 - \epsilon) 2x(pqN)^{1/2} \text{ integers } M_1, M_2, \ldots, M_\ell,
$$

$$
pN - (xpqN)^{1/2} < M_1 < M_2 < \ldots < M < pN + x(pqN)^{1/2} \text{ such that } p_M(Q) > 1 - \epsilon
$$

for every $i, i=1, \ldots, 1$.

ii) If $Q$ is a convex property and $pqN \to \infty$ then a.e. graph in $\mathcal{G}(n,p)$ has $Q$, if for every fixed $x$ a.e. graph in $\mathcal{G}(n,M)$ has $Q$, where $M=pN+x(pqN)^{1/2}$.

The main use of the above theorem is that it allows us to work with the space $\mathcal{G}(n,p)$, which is much easier to work with, and still conclude that a.e. $G_n$ has a certain property $Q$.

When studying $\mathcal{G}(n,M)$ are always takes $M$ to be a function of $n$, with $M=M(n) \to \infty$ as $n \to \infty$, for otherwise a.e. $G_n$ consists of $M$ independent edges; the space $\mathcal{G}(n,p)$ is interesting for constant $p$ as well.

However as shown by the following result, for certain ranges of $p$ and $M$, including the range when $p$ is constant properties given by first order sentences are not too exciting when applied to graphs in $\mathcal{G}(n,M)$ and $\mathcal{G}(n,p)$.

**Theorem 5.1.2.** Suppose $M = M(n)$ and $p = p(n)$ are such that for every $\varepsilon > 0$ we have (1) $Mn^{-2\varepsilon} \to \infty$ and $(N - M)n^{2\varepsilon} \to \infty$, and (2) $pn^\varepsilon \to \infty$ and $qn^\varepsilon \to \infty$.

Let $Q$ be a property of graphs given by a first order sentence. Then either $Q$ holds for a.e. graph in $\mathcal{G}(n,M)$ and $\mathcal{G}(n,p)$ or else it fails for a.e. graph in $\mathcal{G}(n,m)$ and $\mathcal{G}(n,p)$.

The basis of the above theorem is that if a random graph has neither two few nor too many edges then, with high probability, for every pair of small sets of vertices, there is a vertex joined to all vertices in the first set and more from the second. To be precise, if $M = M(n)$ satisfies (1) for every $\varepsilon > 0$ then for every $K \in \mathbb{N}$ a.e. $G_M$ is...
such that whenever $W_1, W_2$ are disjoint $k$-sets of vertices there is a vertex

$\forall \epsilon \in V - W_1 \cup W_2$ joined to every vertex in $W_1$ and to more in $W_2$. A similar assertion holds for $G_p$ if $p = p(n)$ satisfied (2). When studying a property of $G_M$ and $G_p$ given by a first order sentence then $M = M(n)$ and $p = p(n)$ should not be chosen from the ranges given by (1) and (2).

The major discovery of Erdos and Renyi was that most monotone increasing graph properties appear very suddenly. Given a monotone increasing graph property $Q$, $M_0 = M_0(n)$ is a threshold function for $Q$ if

i) a.e. $G_M$ has $Q$ provided $M/M_0 \to \infty$,

ii) a.e. $G_M$ fails to have $Q$ provided $M/M_0 \to 0$.

Infact for many a property $Q$ there is a sharp threshold function: the uncertainty interval around $M_0$ can be very small indeed. Applied to such a property $Q$, all that theorem 5.1.2 says is that $M_0$ is not the range given by (1).

A classical example of a sharp threshold function is the following result.

Theorem 5.1.3 Let $w(n) \to \infty$ and set $M_1 = \left\lfloor \frac{n}{2} \right\rfloor (\log n + (-1)^t w(n))$.

Then a.e. $G_{M_1}$ is disconnected and a.e $G_{M_2}$ is connected.

Definition 5.1.3 As the number of edges $t$ increases from just below $n/2$ to just above $n/2$, the maximal order of a component shoots up from $o(\log n)$ to a positive constant times $n$, is called a giant component and $t = n/2$, the maximum order is about $n^{1/2}$.

We state the following theorem.

Theorem 5.1.4 Let $c > 1$ be a constant and let $t = [(cn/2)]$, $\omega(n) \to \infty$. Then a.e. $G_1$ is such that with the exception of at most $\omega(n)$ vertices, all vertices of $G_1$ belong to the
giant component to components which are trees. Furthermore,

\[ |L_i(G_i) - n \{ 1 - 1/c \sum_{k=1}^{\infty} \frac{(ce^{-c})^k}{k!} \} | \leq o(n)n^{1/2}\log n \]

and if \( k_0 = 1/\alpha \{ \log n - (5/2)\log \log n - l_0 \} \notin N \), where \( \alpha = c - 1 - \log c \) and \( l_0 = o(1) \), then

\[ \max \{ m - 1; L_{m_i}(G_i) \geq k_0 \} \]

has asymptotically poisson distribution with mean

\[ \frac{\alpha^{5/2}}{\lambda - \frac{e^b}{c \sqrt{2\pi} (1-e^\alpha)}} \]

5.2 RANDOM GRAPH PROBLEMS IN POLYMER CHEMISTRY

We examine some random graph problems suggested by polymer chemistry. These problems generalize the random graph that Erdos and Renyi examined. In certain cases the generalization is known to lead to novel graph behaviour and mathematical techniques.

5.2.1 Graph models

Erdos and Renyi[22] considered a random graph on \( n \) labelled vertices, where each of \( M=1/2n(n-1) \) possible edges was independently present (probability \( p \)) or absent (probability \( q=1-p \)). Let \( G \) be an unordered graph on the \( n \) vertices which has \( b \) edges. Then \( G \) is the selected graph with probability \( p^b q^{M-b} \).

Let us generalize the probability measure. Assume \( G \) has partition \( \pi H \). The following constraints on vertex and edge numbers hold.

\[ \sum_n_j = n \quad \text{(1)} \]

\[ \sum j n_j = 2b \quad \text{(2)} \]

Give the graph \( G \) the weight (not probability)
where \{H_j\} and \beta are preassigned numbers. We now select G from all possible graphs with probability

\[
P(G) = \frac{W_0}{\sum W_0},
\]

where the sum runs over all possible graphs satisfying (1).

In equation (3), H_j is the intrinsic tendency of the vertices to be of degree j. The parameter \beta determines the expected number of edges in the random graph.

The Erdos-Renyi study is the specialization

\[
H_j = 1, \quad j=0,1,2,...; \quad \beta = p/q
\]

To extend that study, one could investigate threshold functions and limit theorems for general \{H_j\} as n—\rightarrow \infty.

In the Graph model, ordered graphs present no new features: every unordered graph of partition \pi H_j^n corresponds to exactly \pi(j!)^n ordered graphs. (The half-edges around each vertex of degree j can be numbered \mj! ways). The ordered case with degree weights

\[
H_j = H_j/j!
\]

is equivalent to the unordered case with degree weights H_j.

5.2.2 The Pseudo multigraph model

Polymer chemists have used special cases of this model and the following one extensively. In this section, we assign probabilities according to equations (1) to (4) but now we do so to ordered pseudomultigraph G instead of unordered graphs.
Fig (1) Both unordered pseudographs in (a) have partition $H_2^4$ but they correspond to different numbers of ordered pseudo multigraphs in (b).

Eq.(6) provided an easy passage from unordered graphs to ordered graphs. The figures show that such passage is possible for pseudomultigraph, since the partition does not uniquely define the correspondence between ordered and unordered pseudomultigraphs. There are an infinite number of pseudo multigraphs on $n$ vertices, so one may suspect already that $\beta$, the bonding parameter, must be "small".

The number of distinct ordered pseudomultigraphs of partition $\pi H_j^{n_j}$ is

$$U_{nb} \{\pi H_j^{n_j}\} = n! / \pi n_j! \cdot (2b)! / 2^b b!$$

\textbf{Proof:} Consider the constraint equation (1)

$$\sum n_i = n$$

It gives the number of ways of assigning the degrees to the $n$ labelled vertices

The equation (2) $\sum j n_j = 2b$

\text{gives the number of ordered pseudomultigraphs after the assignment of vertex degrees. If the vertex } V \text{ is to have degree } j, \text{ make } j \text{ copies of the letter } v, \text{ i.e., } V_1, V_2, \ldots, V_j. \text{ There are now } 2b \text{ letters. Permute them } ((2b)! \text{ ways}) \text{ and pair the letters starting from the beginning of the sequence (i.e. position } 2k-1 \text{ is paired with } 2k, k=1,2,\ldots,b). \text{ Join the paired half-edges in the pseudomultigraph. This over counts the}$$
pseudo multigraphs by a factor $2^b b!$ since the order within letter-pairs is immaterial

$(2^b)$ as is the order of the pairs themselves ($b!$)

$$\frac{(2b)!}{2^b b!} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-x^2} dx.$$  \hspace{1cm} (8)

Eqn(7) shows that the exponential generating function of the pseudomultigraph weight is

$$U(v, \beta) = 1 + \sum_{n=1}^{\infty} \frac{v^n}{n! (2b)!} \sum_{n_j} \frac{\pi H_{(n_j)}^j B^n}{n! (2b)!}$$

$$= 1 / \sqrt{2\pi} \int_{-\infty}^{+\infty} \exp \left[ v H (\beta^{1/2} x) \right] e^{-x^2} dx.$$  \hspace{1cm} (9)

where

$$H(x) = \sum_{j=0}^{\infty} H_j x^j.$$  \hspace{1cm} (10)

The second sum in (9) runs over all $\{n_j\}$ satisfying constraints (1) and (2). The second equality results from substitution from (8), reversal of integration and summation and their elementary manipulation of the sum.

Eqn(9) must usually be interpreted formally; the two sides diverge in general.

Taking coefficients of $v^n$ on both sides generally yields a convergent expression for the weights of pseudomultigraphs on $n$ vertices however:

$$U_n(\beta) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} H^n (\beta^{1/2} x) e^{-x^2} dx.$$  \hspace{1cm} (11)

The expected proportion of the $n$ vertices which have degree $j$ is

$$p_j = \frac{1}{n} \frac{H_j}{U_n} \frac{\partial U_n}{\partial H_j}.$$  \hspace{1cm} (12)
Since the effect of the operator $H_j$ is to weight each term of $U_n$ by the number of vertices of degree $j$ present in the corresponding pseudomultigraph.

We assume that $\beta = 1/\nu_n$, where $\nu$ is a fixed constant and $n$ is the number of the expected number of edges in the pseudomultigraph is proportional to $n$, a result desirable to chemists.

Hence $p_j$ is the ratio of two integrals. We apply the method of steepest descents as $n \to \infty$ to find $p_j$:

$$ p_j = \frac{H_j s^j}{H(s)} + o(n) \quad \text{as } n \to \infty \tag{13} $$

where $s$ satisfies the saddle-point condition

$$ \frac{H'(s)}{H(s)} = \nu s \tag{14} $$

The behaviour of the vertex degree distribution as $\nu$ decreases (i.e., as $\beta$ and edge numbers increase) is dependent on the analytic behaviour of $s$. This may or may not be smooth.

If $c(\nu, \beta)$ is the exponential (mv) generating function of the weights $c^k(\nu, \beta)$ for the connected pseudomultigraphs on $n$ vertices, then $U(\nu, \beta)$ of (9) satisfies

$$ U(\nu, \beta) = \exp c(\nu, \beta) = 1 + \sum_{k=1}^{\infty} \frac{c^k(\nu, \beta)}{k!} \tag{15} $$

Heuristic. Every pseudomultigraph can be decomposed into $k$ connected components. $c^k(\nu, \beta)$ is the exponential generating function for ordered $k$-tuples of connected pseudomultigraphs whose vertices are drawn from $n$ labelled points. $1/k!$ removes
k-tuple ordering.

Consider again the random pseudomultigraph on $n$ vertices. Let

$$R_n = \frac{q_n}{n}$$

where $R_n$ is the expected number of vertices of component containing a random vertex. Using eq(15) and standard generating function techniques, we see that $q_n$ obeys the recursion

$$\sum_{k=-m}^{m} T_{m,k} [4k^2 - q_{m-k} - q_{m+k}] = 0 \tag{16}$$

where

$$T_{m,k} = \frac{U_{m+k}(\beta) U_{m+k}(\beta)}{(m-k)!(m+k)!}$$

If $H(x) = e^x$, then Eqn(10) shows

$$U_n(\beta) = e^{1/2\beta x} \tag{17}$$

The asymptotics of $q_n$ for this case by the following novel technique: the term $T_{m,k}$ have maxima amongst the roots of

$$T_{m,k+1} = e^{\beta(2k+1)} = 1$$

$$T_{m,k} = e^{\beta(m+k+1)}$$

For $\beta m \leq 1$ and $m \rightarrow \infty$, Eq(18) has the unique root $k=0$ and $T_{m,k}$ (suitably normalized) approximates a normal density function in $k$ with mean zero and variance $1/2 m(1 - \beta m)^{-1}$. Hence (16) yields

$$q_m = 2E(k^2) = m/(1 - \beta m), \quad \beta m \leq 1 \tag{19}$$

For $\beta m > 1$, $T_{m,k}$ has maxima at the two non-zero roots of Eq(18) which are $o(m)$.

For these $k$,

$$q_{m-k} + q_{m+k} \sim 4k^2, \tag{20}$$

Because $\beta(m-k) \leq 1$ for the positive root of Eq(19) implies $q_{m,k} = o(m)$,
\( \varphi_{m+k} \) is therefore dominated by \( \varphi_{m+k} \) in Eq(20). If \( m+k=N \) and \( \eta = 2k/N \) then \( \beta N > 1 \) and Eq(20) becomes

\[
\varphi_n \sim (\eta N)^2
\]

Eq(18) yields \( e^{N\ln(1-\eta)} = 1 \)

In this model (Whittle's results are consistent with the following as \( \beta \) increases from zero all components are finite trees until \( \beta n > 1 \), when a component of size \( \eta n \) appears. All cycles, loops and multiple bonds remain confined to the giant component. These statements are probably only approximately true as more precise statements have not been investigated yet.

5.2.3 The tree Model

A model for trees similar to those for graphs and pseudomultigraphs using the degree weights \( \{H_j\} \) of sections 5.2.1 and 5.2.2 has not been investigated.

Instead standard models we begin with the distribution of vertex degree \( \{p_j\} \).

\( p_j \) is the probability that a vertex has a given degree. (Eq(12)).

Let \( p(x) \) be the ordinary generating function of \( \{p_j\} \).

\[
p(n) = \sum_{j=0}^{\infty} p_j n^j
\]

Because the correspondence between ordered and unordered trees is a function of tree partition alone (Eq(6)), we examine unordered trees as representative of both cases.

The probability that the vertex on the end of a random edge has degree \( k \) is

\[
f_k = \frac{k \rho_k}{P(1)} \quad k=1,2,3,...
\]

**Proof:** The probability is a priori probability of the vertex being a degree
\(k \{p_k\}\) weighted by the number of edges from the vertex \((k)\). \(1/p'(1)\) normalizes the \(\{f_k\}\) into probabilities.

This effectively specifies the distribution of the trees as a branching process.

\[
\begin{align*}
\text{GEN 0} & \\
\text{GEN 1} & \\
\text{GEN 2} & \\
\end{align*}
\]

Choose a vertex at random Eq(22) gives the distribution of the degree of this "progenitor" vertex. The progenitor's degree is the number of 1-st generation "offspring". (those vertices adjacent to the progenitor). Each offspring vertex in the 1-st generation has degree \(k\) (i.e. \(k-1\) 2-nd generation offspring and 1 parent) independently with probability \(f_k\), as do offspring vertices in subsequent generations.

Results about tree distributions in this model are derived from the corresponding branching process results.

Straightforward extensions of this model are possible: multiple vertex and edge colours directing etc. Perhaps the most interesting extension is to assign to the vertices independently identically chosen random masses \((\omega)\) and to let the vertex
degree probabilities ($P_j$) be functions of $\alpha$. This last notion has obvious extension to the degree weights ($H_j$) of sections 5.2.1 and 5.2.2.

It is likely that an approach to trees through degree weights ($H_j$) would yield results similar to the branching process until a tree containing $o(n)$ of the $n$ vertices formed (this corresponds to a supercritical branching process).

5.3 RANDOM GRAPHS AND POLYMERISATION PROCESSES.

5.3.1 Polymerisation processes with intra-polymer bonding

Whittle [60] has studied clustering processes under the assumptions that new bonds may or may not form within an existing cluster. He showed by two model namely cosh model and poisson model gelation may or may not be accompanied by critical effects at unit level. He has also investigated statistics of polymer size and structure beyond criticality. While dealing with polymerisation processes with intra polymer bonding under several types of unit, he found that poisson interactions were amonable for forming a model for the process.

**Definition 5.3.1** Let $\lambda$ be the intensity of association between a given bonds and $\lambda^i$ when bonding was allowed between units in the same polymers. Given that polymers may be large, and that our level of description affords no measure of distance, it seems proper to regard $\lambda^i$ also as being of order $V^{-1}$(prescribed volume $V$) specifically, we shall define the quantities

$$v = \lambda^i / \lambda, \quad k = \mu / \lambda V$$

and assume then independent of $V$. That is, encounter-rate between units declines as $V^{-4}$ in all circumstances, but is modified by a factor $v$ for intrapolymer as against

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$$v = \lambda^i / \lambda, \quad k = \mu / \lambda V$$

and assume then independent of $V$. That is, encounter-rate between units declines as $V^{-4}$ in all circumstances, but is modified by a factor $v$ for intrapolymer as against
intrapolymer encounters. The quantity $k$ can be regarded as a dissociation rate on a standardised time-scale.

**Definition 5.3.2** Let $R$ denote the size $\sum_{j} r_j$ of an $r$-mer, and $n_r$ denote the number of $r$-mers, so that $N = \sum_{r} R n_r$. We assume $N$ prescribed, and denote the equilibrium probability generating function (p.g.f) of the $n_r$ by

$$H_N(z) = \mathbb{E} \left( \prod_{r} Z_r^{n_r} \right)$$

(1)

We use $C_{n} f(\theta)$ to denote the operation extracting the coefficient of $\theta^n$ in the series expansion of a function $f(\theta)$. This can be given the explicit form

$$C_{n} f(\theta) = \frac{1}{2\pi i} \oint f(\theta) \theta^{-n-1} d\theta$$

(2)

When the integration contour is a tight circuit around the complex $\theta$-origin. We shall also need to define the function,

$$H(\xi) = \sum_{j=0}^{\infty} H_j \xi^j / j!$$

We state the following theorem without proof.

**Theorem 5.3.1.** Suppose the Markov process constituted by the model be irreducible for prescribed $N$, so that the equilibrium distribution is unique. Then

$$H_N(z) \propto C_{n} \left[ \exp \left( C_{r} z^r \right) \right],$$

(3)

Where the function

$$G = \sum_{r} C_{r} \theta^r$$

(4)

has the determination,

$$e^G = \left[ \sqrt{2\gamma / \nu} \int_{-\infty}^{\infty} \exp \left[ V/\nu \left( \theta H(\xi) - (k\xi^2/2) \right) \right] d\xi \right]^{\nu}$$

(5)

Implication of the theorem 5.3.1

**Definition 5.3.3** Approximizing function.
Let us define
\[
P_i = \frac{E(N_i)}{\sum E(N_i)} = \frac{E(N_i)}{N}
\]
as the probability that a unit forms \( j \) bonds and
\[
\varphi(\xi) = \sum p_j \xi^j
\]
as the corresponding p.g.f. Let us also define the operation \( I \) on a function \( f(\xi) \) of \( \xi \) by
\[
I[f(\xi)] = \sqrt{\kappa} V^2 \int f(\xi) e^{\nu(\xi) d \xi},
\]
where
\[
J(\xi) = \varphi \log H(\xi) - \kappa \xi^2 / 2
\]

**Theorem 5.3.2** For the case \( v = 1 \), the following exact results hold:
\[
p_n(H) = V^n I(1)/N! = V^n N! \sqrt{\kappa} V/2 \pi \int \left[ H(\xi)^2 e^{\kappa \xi^2 / 2} \right] d\xi
\]

\[
\varphi(\xi) = \frac{I[H(\xi)]}{I(1)}
\]

\[
E(B) = \frac{N! \left[ \xi H^1(\xi)/H(\xi) \right]}{2I(1)}
\]

For the case \( v = 0 \) the following asymptotic (in the thermodynamic limits) results hold below criticality.
\[
p_n(H) \sim \left( \frac{V}{e} / N \right)^n \exp \left( V J(\bar{\xi}) \right),
\]
\[
\varphi(\xi) \sim H(\bar{\xi}^2)/H(\bar{\xi}),
\]
\[
E(B) = \frac{N \bar{\xi} H(\bar{\xi})}{2H(\bar{\xi})} = \frac{\kappa \bar{\xi}^2}{2}
\]

where \( \bar{\xi} \) is the value of \( \xi \) maximising \( J(\xi) \); this is unique below criticality.

**Definition 5.3.4** The two given quantities of the process are \( H(\bar{\xi}) \) and \( k \); we can
regard then as determining the relative strength of various bond combinations and
the overall strength of bonds respectively. In view of (13) we can regard $H(\xi)$ as being
very nearly the p.g.f of the number of bonds formed by a randomly chosen unit. So it
is reasonable to speak of the binomial case if

$$H(\xi) = (1+\xi)^p,$$

and of the poisson case if

$$H(\xi) = e^\xi.$$

This latter case is an amenable and importance.

Consider now the model for which

$$H(\xi) = \cosh(\xi) = 1/2 (e^\xi + e^{-\xi})$$

$$= \sum_0^{\infty} \frac{\xi^{2j}}{2j!}$$

which we shall term the cash model for obvious reasons. The relative strengths of
bond combinations for this model differ only from those for the poission model in
that units with odd number of bonds are forbidden. The Markov process constituted
by the model is irreducible for a fixed value of $N$, and so has a unique equilibriurm
distribution. However, the case

$$H(\xi) = \cosh(\xi) + \varepsilon \sinh(\xi)$$

$$= \sum_0^{\infty} \frac{\xi^{2j}}{2j!} + \varepsilon \sum_0^{\infty} \frac{\xi^{2j+1}}{(2j+1)!}$$

is perfectly proper in this respect if $\varepsilon > 0$, because transitional structures with
a, of odd $j$ are these possible. Further more (16) shows all the features we wish to
demonstrate, if $\varepsilon$ takes a sufficiently small positive value. So, as it turns out, we can
proceed direct to case (15) without impropriety, and rather more simplicity.

With $H$ given by (15) the quantity $\exp J$ is a mixture of normal densities and is unimodel or bimodel according as $Q \leq k$ or $Q > k$. We can thus expect switching behaviour at $Q = k$. This also turns out to be the gelation point, as emerges latter.

**Definition 5.3.5** Considering the case of poisson interaction

$$H_{\alpha} (\xi) = \exp \left( \sum_{\alpha \beta} t_{\alpha \beta} \xi_{\alpha \beta} \right)$$  \hspace{1cm} (17)

for which the exact evaluation of (5) is possible. Further we can deduce

$$p(H) = \frac{V^N}{\pi N_{\alpha}!} \cdot \frac{1}{2V} \cdot \frac{\exp \left[ - \sum_{\alpha \beta} \frac{t_{\alpha \beta} \phi_{\alpha \beta} N_{\alpha} N_{\beta}}{k_{\alpha \beta}} \right]}{2N_{\alpha} \phi_{\alpha \beta} N_{\alpha} N_{\beta}}$$  \hspace{1cm} (18)

We can evaluate the distribution of $N_1, N_2$ from (18):

$$p(N_1, N_2) \propto \left( \frac{1}{N_1! N_2!} \right) \exp \left[ \left( \phi_1^2 N_1^2 + \phi_2^2 N_2^2 + 2\phi_1 \phi_2 N_1 N_2 \right) \right]$$

$$\frac{1}{2kV}$$

We look for the maximising value, recalling that $N_2 = N - N_1$. We find then that, with stirling approximations to the factorials,

$$\frac{d \log p}{d N_1} = \frac{1}{(D/V)} (N_1 - N_2) \log (N_1 / N_2)$$  \hspace{1cm} (19)

$$\cdot \left( \frac{\phi_1^2 - \phi_2^2}{K} \right)$$

From (19) we see that there is always stationary point at $N_1 = N_2$ and few (20) that this will be a maximum iff
If we set \( N_1 = (QV/2)(1+s) \), then we see from (19) that \( p \) will be stationary for values of \( s \) satisfying

\[
(1-s) e^{-Dp} = (1+s)
\]

This will have a pair of symmetrically placed non-zero roots if equivalence (21) is violated and one readily verifies that \( p \) is then maximal at these non-zero \( s \)-values, and locally minimal at \( s = 0 \).

If \( \varphi_1 < \varphi_2 \) then equidistribution of states is always the stable condition. If \( \varphi_1 > \varphi_2 \) then there is a critical value of density

\[
QC = \frac{(\varphi_1^2 - \varphi_2^2)}{2K}
\]

below which equidistribution is the stable condition, and above which one state or the other predominates. This is evidently the Ising phase transition which would show up also in the distribution of cluster sizes if one examined these.

**Remark 5.3.1** Criticality as revealed by unit distribution over regions. If a value of \( \xi \) maximises \( J(\xi) \) globally we shall refer to it as a stable solution if it maximises \( J(\xi) \) locally we shall refer to it as a locally stable solution; if it is locally stable but not stable we shall refer to it as measurable. These characterisations are supported by the fact that maxima of \( J \) corresponds to maxima of the same character of \( p(N_1, N_2, \ldots, N_m) \).

**Theorem 5.3.3.** The necessary and sufficient conditions for a value \( \xi \) to maximise \( J(\xi) \) locally are that should satisfy

\[
\theta H_1(\xi) = k_\xi \xi,
\]
where
\[ \theta = \frac{\varrho}{\tilde{H}} = \varrho\left( \sum_i \frac{H_i(\xi_i)}{i} \right) \]
and that either (1) the smaller root of (22) be taken for all \( i \) or (ii) the smaller root of (22) be taken for all but one value of \( i \), and
\[ S \triangleq 1 + \sum \frac{f_i(H_i/H)^2}{(k_i/q)-(H_i^{11}/H)} \leq 0 \quad (23) \]

Remark 5.3.2 Polymer size

For simplicity we consider only the homogeneous case, for which
\[ p(H) = C_N(e^{\xi_0}) \]
reduces to
\[ P_N(H) \propto C_N(I^m) = C_N(e^{\alpha G}) \]
in the case \( \nu = 1 \). Here
\[ I = e^\alpha = \sqrt{kV/2\pi m} \int \rightarrow \exp[V/m(0H(\xi) - k\xi^2/2)]d\xi \]
The average polymer size \( R \) has the expression
\[ R = \frac{C_N(G_{\alpha I}^m)}{C_N(G_{\alpha I}^m)} = \frac{C_N(I_{\alpha I}^{m-1} - I_{\alpha I}^{m-2})}{C_N(I_{\alpha I}^{m-1})} \]
where the subscripts \( \alpha \) indicate differentiation with respect to \( \alpha = \log \theta \).

Theorem 5.3.4 The average polymer size has the expression
\[ \bar{R} = 1 + \frac{\sum H_k}{\overrightarrow{J}[\sum \frac{(H_1 - H_2)^2}{2}]/\overrightarrow{J}(1)} \]
(Where \( \overrightarrow{J} \) denotes the weighted average operator)
with asymptotic evaluations
\[ \bar{R} \sim \frac{Q[H,\omega)/H(\omega)]^2}{k- QH^{11}(\omega)/H(\omega)} \]

below criticality and

\[ \bar{R} \sim N \left[ \frac{H(\omega_1)-H(\omega_2)}{H(\omega_1)+(m-1)H(\omega_2)} \right]^2 \]

above criticality. Here \( \xi(\omega_1, \omega_1, \ldots, \omega_1) \) and \( \xi(\omega_2, \omega_2, \ldots, \omega_2) \) are subcritical and supercritical solutions of (22).

5.4. NATURE OF STATISTICS OF POLYMERS.

5.4.1 Polymer bonding

A graph on \( N \) labelled nodes can be regarded as a pattern of bonding between \( N \) distinguishable particles, or units. It thus provides a representation of a pattern of interaction between these units in which bonding or its absence is the only relationship, notions such as physical distance or interaction as a function of distance being absent. Thus skeletal version of interaction is that generally adopted in the literature on polymerisation: the study of the molecules formed by bond-formation between units.

The nodes of the graph represent units, and are supposed distinguishable i.e. labelled. The modes of the graph may be coloured if different types of unit are possible.

The arcs of the graph represent bonds. If the arcs are directed then they indicate a directional property of the bond; perhaps that of being initiated by unit from which the arc emerges. Multiple bonding and self-bonding are not excluded in general.
The connected components of the graph represent molecules, or polymers. Suppose that one allows the bonding pattern to be random, so that one is effectively considering random graphs; i.e. a probability distribution over the set of possible graphs on the N assigned nodes. Then there is considerable interest in the nature of the statistics of the polymers induced by the prescribed statistics on bonding. This interest has been followed in both the mathematical and the physical literature i.e., in the literature on random graphs and in the literature on polymerisation.

The configuration of the graph is described completely by the pattern of directed arcs between the N labelled nodes. That is, by \( s = \{ s_{ab} \} \) where \( s_{ab} \) is the number of arcs from node a to node b (a,b = 1, 2, ..., N). By \( \sum \) we indicates a summation over all configurations for prescribed N; by \( \sum \) we indicate a summation over \( N = 1,2,......... \).

**Lemma 5.4.1** Suppose that to each graph \( \mathcal{C} \) 'on an arbitrary number of labelled nodes can be attached a weight \( w(\mathcal{C}) \) with the properties: (1) that \( w(\mathcal{C}) \) is invariant under permutation of the nodes, and (ii) that \( w(\mathcal{C}) = w(\mathcal{C}_1)w(\mathcal{C}_2) \) if \( \mathcal{C} \) can be decomposed into mutually unconnected graphs \( \mathcal{C}_1, \mathcal{C}_2 \).

Then
\[
\sum_{\mathcal{C}} \frac{w(\mathcal{C})}{N!} = \exp[\sum w(\mathcal{C})] 
\] .......(1)

where the sum \( \sum \) covers all distinct connected graphs, \( N \) is the number of nodes of \( \mathcal{C} \), and the term for \( N = 0 \) is taken as unity.

Suppose that \( p_N(s) \) is the probability of configuration \( s \) on the given \( N \) nodes, and that
\[
p_N(s) \propto Q_N(s) := \prod_i \psi_{i}^{m_i} 
\] .......(2)
where \( i \) indexes the possible types of connected graph. (i.e polymer at the completest level of prescription) and \( m_i \) is the number of times component \( i \) occurs in the graph. The proportionality constant is (2) will be a function of \( N \) alone which normalises the distribution over \( s \) for fixed \( N \). The weighting \( Q_N(s) \) evidently satisfies the conditions of the lemma, and identity (1) then becomes.

\[
\sum_N \sum_s Q_N(s)/N! = \exp(\sum_i \psi_i)
\]

Now, typically, one will classify polymers of some lower level of description, indexed by \( r \), say, we shall refer to a polymer of type \( r \) as an \( r \)-mer. Presumably the description will also determine the number of units in the \( r \)-mer; let us denote this by \( R \). Let \( r(i) \) and \( R(i) \) denote the polymer type and size of component \( i \) and let us define \( g_r = \sum_i \psi_i \), \( r(i) = r \).

Let us denote the number or \( r \)-mer in the configuration by \( n_r \), and define the probability generating function

\[
\pi_n(z) = E(\pi z_r^n)
\]

This defines the polymer statistics, which we assume to be the object of interest.

**Theorem 5.4.1** Assume the distribution of complete configurations given by (2).

Then \( \pi_n(z) \) is proportional to the coefficient of \( \theta^N \) in the expansion of

\[
\exp[\sum_i \theta^{R(i)} \psi_i z_r^{m_i} ] = \exp[\sum_r \theta^R g_r z]
\]

Proof: We take the identity (1) with

\[
W(\bar{Q}) = Q_N(s) \theta^N \pi z_n^{n_r} = \pi [\theta^{R(i)} \psi_i z_r^{m_i} ]
\]

and obtain

\[
\sum_N \theta^N! [\sum_s Q_N(s) \pi z_r^{n_r}] = \exp(\sum_r \theta^R g_r z_r)
\]

The coefficient of \( \theta^N \) in the left-hand-member is indeed proportional to \( \pi_n(z) \)
It is not necessary that either of the infinite series in (3) should converge for any θ. It is simply asserted that coefficients of θ^N on the two sides are equal for all N. However, if the series do converge for some θ, then the identity has an interesting interpretation. It asserts that there is an open version of the process (i.e. a version in which N is also a random variable) in which n_r are independent poisson variables. The two conditions of Lemma 5.4.1, which lead to this conclusion, are: (1) the units are statistically identical, even if distinguishable, and (ii) the appearances of polymers would be independent events were it not for constraints on N.

The solution for π_N(θ) yielded by Theorem 5.4.1 is moderately explicit if the coefficients g_r can in fact be calculated. What can indeed often be calculated in the sum

\[ G(θ) = \sum_r θ^R g_r = \sum_i θ^{R(i)} \psi_i. \]

If we set all the z_r equal to unity in identity (3) then this reduces to

\[ \sum_N \frac{θ^N Q_N}{N!} = e^{G(0)} \]

where

\[ Q_N := \sum_s Q_N(s) \]

and these expressions are often computable. It may then be that g_r can be calculated from G(θ) if the \( \psi_i \)'s include variables which can serve as marker variables. Indeed, the appropriate level of description of the polymers is fixed by the level at which such marker variables exist, and g_r is extractable from G(θ). Otherwise expressed, one distinguishes just those polymers which are energetically distinguishable.
Suppose that we choose
\[
Q_N(s) = V^N \pi \frac{(\delta / 2)^{s_{ab}}}{a \ b \ s_{ab}!}
\]

Here \(V\) is the "Volume" of the region within which the \(N\) units are constrained, and we assume that
\[
\delta = \frac{\lambda}{V},
\]

where \(\lambda\) is a parameter measuring intensity of bonding. The factor \(V^N\) included in (5) is constant in that it is independent of \(s\), but it becomes important when we consider a distribution of polymers over more than one "compartment" in space.

The assumption implicit in (5) is that the \(N^2\) quantities \(s_{ab}\) are independent Poisson variables, each with expectation \(\delta / 2\). Later we shall consider the thermodynamic limit, in which \(N\) and \(V\) become infinite, the density \(Q = N/V\) being held fixed. The point of the dependence (6) is then that the total expected number of bonds issuing from a prescribed unit remains fixed at \(N\delta = \lambda Q\).

This Poisson assumption seems the simplest possible: polymer statistics are purely combinatorial in that all polymers with \(R\) units and \(L\) bonds have the same "energy". These two statistics then constitute the natural description level for polymers in this model. The Poisson assumption leads to more natural mathematic than the assumptions of independently occurring single undirected bonds. The assumption allows the phenomena of self bonding and multiple bonding, which may seem unnatural. It is not clear that they are, but their occurrence has negligible effect in the thermodynamic limit.
We have

\[ Q_N = V^N e^{\lambda N/V^2} \]  

so that

\[ e^{G(0)} = \sum_{N=0}^{\infty} \frac{(\theta V)^N}{N!} e^{\lambda N/V^2} \]  

The series (8) defines \( G(0) \) as a power series in \( \theta V \) and \( \lambda / V \),

\[ G(\theta) = \sum_{R} \sum_{L} w_{RL} \theta^R (\lambda / V)^L \]

\[ = \sum_{R} \sum_{L} w_{RL} V^{R-L} \lambda^L \theta^R \]

where \( w_{RL} \) is a purely combinatorial term. The pair \( R, L \) constitutes the natural prescription \( r \) at this level, with \( \theta \) and \( \lambda \) being the marker variables for \( R \) and \( L \) respectively. We have the identification

\[ g_{RL} = w_{RL} V^{R-L} \lambda^L \]

Note now that

\[ R-L \leq 1 \]

with equivalence if and only if the polymer is a tree. We see then from (10) that tree polymers have an abundance of order \( V \) while other polymers (let us call them ring-polymers) have an abundance of lower order, the order decreasing with the amount of cyclisation of the polymer. One can say that there are so many ring-polymers that the abundance of any given one must be low, although their total effect may be appreciable.

Note that the series (8) always diverges, as must then the series (9). This is because of the multitude of contribution from ring-polymers. If one considers the contributions to these series from tree polymers alone then the series converge so long as \( \theta \) does not exceed a value which is related to the phenomenon of gelation.
As indicated in [6i] by far the easiest way to detect gelation is to examine the quantity $P_{N+n} P_{N-n}$ as a function of $n$ where $P_{N} = Q_{N}/N!$. This quantity is the probability that $2N$ units should distribute themselves over two disjoint regions each of volume $V$ as $(N+n, N-n)$, bonding being allowed only within each volume according to the prescription (5). In the sol-state $P_{N+n} P_{N-n}$ has its maximum at $n=0$ (corresponding to statistical equi-distribution between the two regions): in the gel-state it has symmetric maxima at $n = \pm n_0$ say (corresponding to a tendency for the units to dump in one region or the other). Using then the criterion

$$P_{N+1} P_{N-1} \geq P_N^2 \quad (11)$$

as a criterion for gelation and the evaluation (7); we find that the condition for gelation in the thermodynamic limit is

$$\lambda Q \geq 1 \quad (12)$$

This agrees with the value deduced by much less direct arguments.

5.4.3 Bond interactions

Let a unit which has formed $j$ bonds (in either directions) be denoted by $a_j$ and let $n_j$ be the number of $a_j$'s in the $N$ given units ($j=0,1,2,...$). Suppose the poisson specification (5) now modified to

$$Q_N(s) = V^N [\prod_{a \neq b} \frac{s_{ab}}{s_{ab}!} ] \pi H_j^{n_j} \quad (13)$$

The assumption is then that the basic poisson statistics are modified by bond-number dependent terms, the term $H_j$ being high or low according as a unit with $j$ bonds is
relatively favoured.

Alternatively expressed, the energy of the configuration is a function only of the bond-numbers $j$, and is low or high according as $H_j$ is high or low.

Now $Q_N$ has the evaluation [8]

$$Q_N = V_n^{1/2} \pi \delta^2 \int_{-\infty}^{\infty} H(\xi)^N e^{\xi/\sigma} d\xi$$

(14)

where

$$H(\xi) = \sum H_j \xi^j/j!$$

The natural description $r$ of a polymer is now $r=(r_0,r_1,r_2,...)$ where $r_j$ is the number of $a_j$'s the $r$-mer contains. The $H_i$ are natural marker variables for the $r_j$.

So still are $\theta$ and $\delta$ for $R$ and $L$, although these are unnecessary in that the $r_j$ determine $R$ and $L$.

$$R = \sum_{j} a_j$$

$$L = 1/2 \sum_{j} ja_j$$

The series (4) with the evaluation (14) for $Q_N$ determines $G(0)$ as a power series in $\theta V$, $\lambda / N$ and the $H_j$

$$G(0) = \sum_{r} w_r V^{r-1} \lambda^{r} \theta^r (\pi H_j^f)$$

where $w_r$ is purely combinatorial, and we have

$$g_r = w_r V^{r-1} \lambda^{r} (\pi H_j^f)$$

for gelation, where $\xi$ is determined by

$$H(\xi) = \sum H_j \xi^j/j!$$

for gelation, where $\xi$ is determined by

$$\lambda \xi \quad = \quad 1$$

$$H(\xi)$$
All this work can be generalised naturally to the case of several unit-types, and to the case when there is a differential effect between inter-and intra-polymer bonding an effect which encourages or discourages cyclisation[62,63]

5.4.4 Equilibrium solutions as time-dependent solutions

Flory's [24] original approach was not to consider the equilibrium version of association and dissociation, but rather to consider the time-dependent version of purely associative process. That is, a process in which bonds form but do not break. He then deduced the occurrence of gelation at a time when the degree of bonding had reached the critical value.

This view expresses the experience of the polymer chemist: when physical conditions are created which favour polymerisation, then polymerisation proceeds progressively, and after some time, gelation is observed as a rather definite event.

Flory's mathematical approach was a mixture of statistical mechanics and heuristics. However one is impelled to ask whether equilibrium distributions such as (5) and (13) might not be seen as time-dependent distribution for an appropriate stochastic process of pure bonding.

Consider time-dependent version of (5)

\[
Q(s,t) = \frac{(\lambda t/2V)^{s_{ab}}}{s_{ab}!} \frac{V^n \pi \prod_{a,b} \xi_{s_{ab}}}{s_{ab}!}
\]  

(15)

Here the bonding parameter \( \delta = \lambda /V \) has been replaced by \( \lambda t/V \). We have also dropped the \( N \) subscript for simplicity.

Distribution (15) implies that the \( s_{ab} \) are independent poisson variables with expectation \( \lambda t/2V \) at time \( t \). This is exactly the distribution that would hold for a
process, completely dissociated at \( t=0 \), in which for any \( a,b \) there is a fixed probability intensity \( \lambda/2V \) that a new bond will form unit \( a \) to unit \( b \), independent of the previous history of the process. This is the simplest stochastic bonding process one can imagine, and one sees from (12) that it gets at time

\[
t = 1/\lambda_Q
\]

Let us write the transition in which \( s_{ab} \) is increased by one as \( s \rightarrow s+e_{ab} \), and denote the corresponding transition intensity as \( \Lambda(s,s+e_{ab}) \). Then the poisson model we have just considered is characterised by

\[
\Lambda(s,s+e_{ab}) = \lambda/2V \quad (a,b = 1,2, \ldots N)
\]

these being the only transitions possible.

Consider now the more general distribution

\[
Q(s,t) = \left( \frac{\lambda t}{2V} \right)^{s_{ab}} \prod_{a b} \frac{\pi}{s_{ab}!} \psi(s) \tag{16}
\]

in which the time-dependent poisson statistics are modified by a time-dependent term \( \varphi(s) \), representing interaction effects associated with the potential energy of configuration \( s \). This can indeed be seen as the time-dependent solution, either exactly or in the thermodynamic limit, of a natural stochastic bonding process.

**Theorem 5.4.2:** Consider the pure bonding process for which

\[
\Lambda(s,s+e_{ab}) = \frac{\lambda \varphi(s+e_{ab})}{2V \varphi(s)}
\]

and let \( p(s,t) \) denote the distribution of \( s \) at time \( t \) for this process, given complete dissociation at time \( t = 0 \). Let
denote the normalised version of expression (16). Then \( p, p^* \) respectively satisfy.

\[
\frac{\partial p(s,t)}{\partial t} = \sum \sum p(s-e_{ab}, t) \wedge (s-e_{ab}, s) - \wedge (s) p(s,t)
\]

\[
\frac{\partial p^*(s,t)}{\partial t} = \sum \sum p^*(s-e_{ab}, t) \wedge (s - e_{ab}, s) - \wedge^* (t) p^*(s,t)
\]

where

\[
\wedge (s) := \sum \sum \wedge (s, s+e_{ab}) = \frac{1}{2V\varphi(s)} \sum \sum \varphi(s+e_{ab})
\]

\[
\wedge^* (t) = \sum p^*(s, H \wedge(t))
\]

If for every \( t \) \( \wedge(s) \) is constant (in \( s \)) for all \( s \) such that \( p^*(s,t) > 0 \) then \( p(s,t) \equiv p^*(s,t) \).