

# A graph-theoretical approach to statistics and dynamics of tree-like molecules

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We present a new graph-theoretical method for calculating the dynamical and statistical properties of a Gaussian chain with various molecular architecture. The characteristic polynomial for its line graph, which has the bonds of a molecular graph as its beads with adjacency of bonds as in the graph, makes it possible to provide us with the general equation for calculating the radius of gyration of Gaussian chains and their relaxation spectra.

## 1. Introduction

The Gaussian chains have played a central role in studies on the statistics and dynamics of flexible chain molecules. A Gaussian chain is a molecule represented as a collection of beads being linked together with Hookean springs. According to the central limit theorem in statistical physics, the random-flight statistics of a flexible polymer can be described by a Gaussian chain which is mathematically simpler to handle [13,16]. Rouse [32], Bueche [6], and Zimm [41] demonstrated that the dynamics of dilute solution of linear polymers can be characterized by considering a Gaussian chain suspended in a flowing viscous liquid. Subsequently, the application of this model to any branched molecule was made by Ham [18] and Zimm and Kilb [42].

The mathematical representation of the graph theory enables us to generalize the statistics and dynamics of Gaussian chains to include any type of branching [12]. Recently, application of concepts of the graph theory to polymer chemistry has been increasing [4,10,14,25,28]. In this work, we present a graph-theoretical method for calculating high-order moments of the radius of gyration and relaxation spectra of flexible chain molecules with any type of branching. The central feature of this approach lies in the statistics and dynamics of chain molecules being reformulated into a more convenient algebraic form.

## 2. Chain dynamics

In this work, we adopt any branched Gaussian chain containing no loops or circles with  $N$  beads and  $N - 1$  segments acting as Hookean springs. Let the spring constant of the bond be  $dkT/b^2$ , where  $k$  is the Boltzmann constant,  $T$  is the temperature,  $d$  is the space dimension, and  $b$  is the effective bond length. A Gaussian chain model for a branched molecule is presented in figure 1(a). Considering a Gaussian chain suspended in a flow liquid, the motion equation is given by

$$-\zeta_0 \dot{\mathbf{r}} = \frac{dkT}{b^2} \mathbf{Z} \mathbf{r}, \quad (1)$$

where  $\mathbf{r}$  is a  $d \times N$  matrix whose rows contain the dimensional component of the  $N$  position vectors of beads,  $\dot{\mathbf{r}}$  is the time derivative of  $\mathbf{r}$ ,  $\zeta_0$  is the friction constant of the beads, and  $\mathbf{Z}$  is the  $N \times N$  connectivity matrix which is called the Zimm matrix in the polymer physics [41] and is known as the Kirchhoff matrix [26] in the graph theory. According to the Rouse theory [32], equation (1) can be rewritten in terms of bond vectors  $\mathbf{b}_i = \mathbf{r}_{i+1} - \mathbf{r}_i$  as

$$-\zeta_0 \dot{\mathbf{b}} = \frac{dkT}{b^2} \mathbf{R} \mathbf{b}, \quad (2)$$

where  $\mathbf{b}$  is a  $d \times (N - 1)$  matrix whose rows contain the dimensional component of the  $N - 1$  bond vectors. The matrix  $\mathbf{R}$  is the  $(N - 1) \times (N - 1)$  connectivity matrix which is called the Rouse matrix [32]. Denoting by  $\Phi(\mathbf{A}; \lambda)$  the characteristic polynomial  $\text{Det}[\mathbf{A} - \lambda \mathbf{E}]$  of a matrix  $\mathbf{A}$ , where  $\mathbf{E}$  is the identity matrix, we can find the following relation:

$$\Phi(\mathbf{Z}; \lambda) = \lambda \Phi(\mathbf{R}; \lambda). \quad (3)$$

The eigenvalues of  $\mathbf{Z}$  contain one zero eigenvalue and, hence, do not possess an ordinary inverse. Equation (3) shows that the non-zero eigenvalues of  $\mathbf{Z}$  are identical with those of  $\mathbf{R}$ . The zero eigenvalue of  $\mathbf{Z}$  represents the mode of chain translation [17]. The springs and beads are assigned in any arbitrary fashion, for example, as shown in figure 1(a), so that the entries of connectivity matrices  $\mathbf{Z}$  and  $\mathbf{R}$  are dependent on how the springs and beads are numbered. However, the eigenvalues of  $\mathbf{Z}$  and  $\mathbf{R}$  are independent of how their elements are numbered [12,14,20]. Each eigenvalue  $\lambda_i$  ( $i = 1, 2, \dots, N - 1$ ) of  $\mathbf{R}$  or non-zero eigenvalue of  $\mathbf{Z}$  is associated with the relaxation times  $\tau_i$  of the  $i$ th mode [7,29]:

$$\tau_i = \frac{\zeta_0 b^2}{2dkT} \lambda_i^{-1}. \quad (4)$$

In the graph theory, a chain molecule is represented as an ordinary graph  $G$ , in which a bead (or vertex) and a segment bond (or edge) are arbitrarily numbered, and a digraph  $D$ , in which each edge of the graph  $G$  is arbitrarily directed, as exemplified in figure 1(b) and (c). The adjacency matrix  $\mathbf{A}$  of the graph  $G$  or the digraph  $D$  has been useful for characterizing and encoding the skeletal structure of molecules [9,24,35,37].

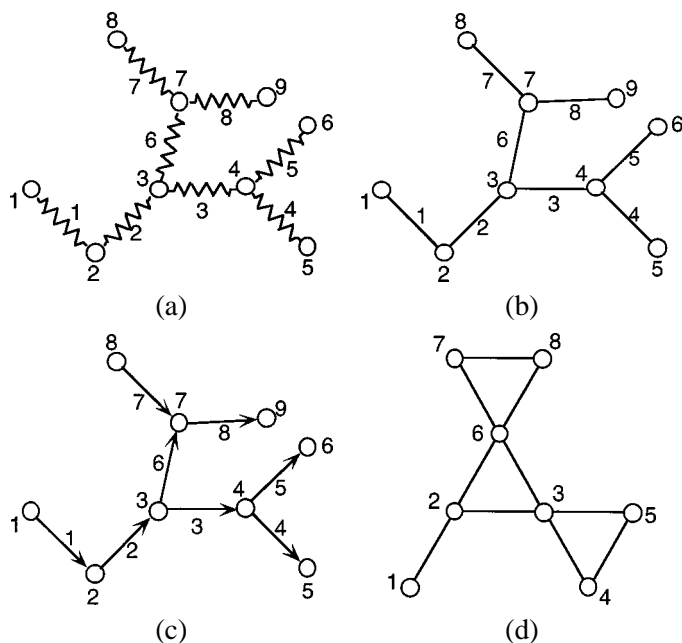


Figure 1. Representation of a flexible randomly coiled chain by (a) the bead–spring model (Gaussian chain model); (b) the ordinary graph; (c) the digraph; (d) the line graph. The beads and springs, lines, or arrows are arbitrarily assigned. The line graph (d) is transformed from the graph (c).

The entry in the  $i, j$  cell of  $\mathbf{A}$  is 1 if there is an edge (or directed line) of  $G$  (or  $D$ ) from a vertex  $i$  to a vertex  $j$ , and this entry is 0 otherwise [5,20].

The Zimm matrix  $\mathbf{Z}$  or the Rouse matrix  $\mathbf{R}$  can also be constructed in a different manner by making use of the incident matrix of a digraph  $D$ . The elements of the incident matrix  $\mathbf{B} = (b_{ij})$  of a digraph are defined by [5,20]

$$b_{ij} = \begin{cases} +1 & \text{if edge } j \text{ starts from vertex } i, \\ -1 & \text{if edge } j \text{ terminates in vertex } i, \\ 0 & \text{otherwise.} \end{cases} \quad (5)$$

According to Forsman [12],  $\mathbf{Z}$  and  $\mathbf{R}$  are given by

$$\mathbf{Z} = \mathbf{B}\mathbf{B}^T \quad (6)$$

and

$$\mathbf{R} = \mathbf{B}^T\mathbf{B}, \quad (7)$$

where the superscript “T” indicates the transpose of a matrix.

Now we consider an incidence matrix  $\mathbf{C}$  of the molecular (undirected) graph  $G$  with elements of  $\mathbf{C} = (c_{ij})$  given by [5,20]

$$c_{ij} = \begin{cases} +1 & \text{if edge } j \text{ is connected with vertex } i, \\ 0 & \text{otherwise.} \end{cases} \quad (8)$$

Introducing a connectivity matrix  $\mathbf{K}$  formed by

$$\mathbf{K} = \mathbf{C}^T \mathbf{C}, \quad (9)$$

we can derive the following relation as shown in appendix A:

$$\Phi(\mathbf{R}; \lambda) = \Phi(\mathbf{K}; \lambda). \quad (10)$$

It should be noted that all elements of  $\mathbf{K}$  are the absolute values of those of  $\mathbf{R}$ . Furthermore, according to a theorem in the graph theory, the following relation has the form for tree graphs [5]:

$$\mathbf{K} = \mathbf{A}_L + 2\mathbf{E}, \quad (11)$$

where  $\mathbf{A}_L$  is the adjacency matrix for the line graph  $L(G)^1$  of  $G$ . The elements of  $\mathbf{A}_L$  are given by [5,36]

$$a_{ij} = \begin{cases} +1 & \text{if edge } j \text{ is connected with edge } i, \\ 0 & \text{otherwise.} \end{cases} \quad (12)$$

An example of the line graph is shown in figure 1(d).

As can be seen from equation (4), the relaxation spectrum of a Gaussian chain can be determined from the eigenvalue spectrum of the Rouse matrix  $\mathbf{R}$ . A combination of equations (10) and (11) gives

$$\Phi(\mathbf{R}; \lambda) = \Phi(\mathbf{A}_L; \lambda - 2). \quad (13)$$

Consequently, the eigenvalues  $\lambda_i$  of  $\mathbf{R}$  can be related to the eigenvalues  $\mu_i$  of  $\mathbf{A}_L$  as follows:

$$\lambda_i = \mu_i + 2. \quad (14)$$

Remembering equation (4), we get

$$\tau_i = \frac{\zeta_0 b^2}{2dkT} (\mu_i + 2)^{-1}. \quad (15)$$

It follows that the relaxation spectrum of a chain molecule is determined entirely by the set of eigenvalues of the adjacency matrix  $\mathbf{A}_L$  of its line graph.

### 3. Chain statistics

The potential energy of the chain molecule can be written as

$$V = \frac{dkT}{2b^2} \text{Tr}[\mathbf{rZr}^T], \quad (16)$$

<sup>1</sup> The line graph  $L(G)$  of  $G$  is the graph having one vertex corresponding to each edge of  $G$  and such that two vertices of  $L(G)$  are joined if and only if the corresponding edges of  $G$  are incident.

where  $\text{Tr}$  denotes the trace of a matrix. Fixman [11] showed that the distribution function of the square radius of gyration  $S^2$  is given by

$$P(S^2) \propto \int \delta(S^2 - N^{-1} \text{Tr}(\mathbf{r}\mathbf{r}^T)) e^{-V/kT} d\mathbf{r}, \quad (17)$$

where  $\delta(x)$  is a Dirac delta function. Introducing a Laplace transform of  $P(S^2)$  with respect to  $S^2$ , we obtain

$$\mathcal{L}[P(S^2)] = \int_0^\infty e^{-zS^2} P(S^2) dS^2 = \varphi(z)^{-d/2} \quad (18)$$

and

$$\varphi(z) = \text{Det}[\mathbf{E} + \gamma z \mathbf{R}^{-1}], \quad (19)$$

where  $\gamma = 2b^2/(dN)$ . Details of the precise derivation of equation (18) are presented in appendix B. The Laplace transform of  $P(S^2)$ , i.e., the generating function of  $P(S^2)$ , provides the average  $\langle e^{-zS^2} \rangle$  and, therefore, the average of powers of  $S^2$  can be computed by making use of the expansion in the form of a power series in  $z$ . Thus we have

$$\langle S^{2n} \rangle = (-1)^n \left. \frac{\partial^n}{\partial z^n} \varphi(z)^{-d/2} \right|_{z \rightarrow 0}. \quad (20)$$

Since we have  $\text{Det}|\mathbf{R}| = N$  for any tree graph as shown in appendix A, equation (19) can be rewritten as

$$\varphi(z) = \frac{1}{N} \text{Det}[\mathbf{R} + \gamma z \mathbf{E}] = \frac{1}{N} \Phi(\mathbf{R}; -\gamma z). \quad (21)$$

Using equation (13), we have

$$\varphi(z) = \frac{1}{N} \text{Det}[\mathbf{A}_L + (\gamma z + 2)\mathbf{E}] = \frac{1}{N} \Phi(\mathbf{A}_L; -\gamma z - 2). \quad (22)$$

It was shown that the characteristic polynomial of the line graph gives the general equation for calculating the radius of gyration of a Gaussian chain with any type of branching.

#### 4. Applications

The coefficient of  $z$  in the characteristic polynomial  $\phi(z)$  can be related to a topological index [3], i.e., the Wiener index [40] of the molecular graph  $G$ , i.e., a total sum of the elements of its distance matrix [5], which is potentially useful in the correlation of molecular topology to thermodynamic properties for alkanes. Therefore, any high-order coefficient in  $\phi(z)$  for a tree-like graph, or any coefficient of the characteristic polynomial for its line graph, has a potential to be a new topological index [24] which can be related to other physical and chemical properties. In this section, the

relationship between the characteristic polynomial and mechanical relaxation processes is examined.

The characteristic polynomial  $\phi(z)$  can be rewritten in terms of the  $N - 1$  eigenvalues of the Rouse matrix as [11]

$$\varphi(z) = \prod_{i=1}^{N-1} \left(1 + \frac{\gamma}{\lambda_i} z\right). \quad (23)$$

Making use of the relation

$$\ln \varphi(z) = \sum_{i=1}^{N-1} \ln \left(1 + \frac{\gamma}{\lambda_i} z\right) = \sum_{i=1}^{N-1} \left[ \frac{\gamma}{\lambda_i} z - \frac{1}{2} \left(\frac{\gamma}{\lambda_i}\right)^2 z^2 + \frac{1}{3} \left(\frac{\gamma}{\lambda_i}\right)^3 z^3 - \dots \right], \quad (24)$$

the sums of reciprocal powers of the eigenvalues are easily determined from the following equation:

$$(n-1)! \sum_{i=1}^{N-1} \left(\frac{\gamma}{\lambda_i}\right)^n = (-1)^{n-1} \frac{\partial^n}{\partial z^n} \ln \varphi(z) \Big|_{z \rightarrow 0}. \quad (25)$$

Using equation (4), we have

$$\sum_{i=1}^{N-1} \tau_i^n = \frac{(-1)^{n-1}}{(n-1)!} \left(\frac{N\zeta_0}{4kT}\right)^n \frac{\partial^n}{\partial z^n} \ln \varphi(z) \Big|_{z \rightarrow 0}. \quad (26)$$

According to the theory of linear viscoelasticity [15], the relaxation spectrum,  $H(\tau)$ , is given by

$$H(\tau) = \frac{ckT}{N} \sum_{i=1}^{N-1} \delta(\ln \tau - \ln \tau_i), \quad (27)$$

where  $c$  is the concentration of beads per unit volume. Then the zero-shear-rate viscosity  $\eta_0$  and the steady-state compliance  $J_e^0$ , a measure of the elastic energy stored under steady flow, are

$$\eta_0 = \frac{ckT}{N} \sum_{i=1}^{N-1} \tau_i, \quad J_e^0 = \frac{N}{ckT} \left( \frac{\sum_{i=1}^{N-1} \tau_i^2}{\left(\sum_{i=1}^{N-1} \tau_i\right)^2} \right). \quad (28)$$

Consequently, the expressions for these two fundamental viscoelastic properties can be derived from equation (26):

$$\eta_0 = \frac{c\zeta_0}{4} \dot{\varphi}_0, \quad (29)$$

$$J_e^0 = \frac{N}{ckT} \left(1 - \frac{\ddot{\varphi}_0}{\dot{\varphi}_0^2}\right), \quad (30)$$

where the suffix 0 indicates the limiting values when  $z \rightarrow 0$ . It was shown that these rheological parameters of any branched polymer can be computed from the characteristic polynomial of its line graph.

## 5. Remarks

In the graph theory, the adjacency matrix is an important device for characterizing the topological feature of graphs, and the algebraic properties of the characteristic polynomials have been extensively examined [2,19,30,33,38]. In this work, we have established the relation between the graph theory and the Rouse theory. Thus, the problems of the dynamics and statistics of a tree-shaped molecule were found to be completely reduced to the eigenvalue problem of the adjacency matrix of its line graph  $L(G)$ . This suggests that various ideas and concepts thus obtained from the graph theory can be introduced directly to polymer physics and chemistry through the use of the adjacency matrix of the line graph.

The significance of the present graph-theoretical approach is to provide the general equations for the relaxation spectrum and the radius of gyration of any tree-like chain. In particular, it is noteworthy that the mathematical method has the potential to provide an algorithmic way to calculate high-order moments of the radius of gyration and the relaxation time for any tree-like chain. These values can be hardly calculated from the usual statistical methods because of a great difficulty in the numeration of the distribution function.

So far, the characteristic polynomial of the adjacency matrix of the graph  $G$  has been considered to be capable of expressing the physical and chemical behaviors for any molecule. Particular interest, therefore, has been focused on the existence of isospectral tree graphs since a pioneering work by Collatz and Sinogowitz [8]. The isospectral graphs [1,2,8,21–23,27,30,31,34] are topologically nonidentical (or isomorphic<sup>2</sup>) graphs whose nonidentical adjacency matrices give an identical polynomial and an identical set of eigenvalues. It has been long identified that the graph-theoretical statement that some molecules have isospectral mates presents a difficulty in discriminating their topological structure.

The present result strongly suggests that the adjacency matrix of their line graph  $L(G)$  plays an important role in the correlations of topological structure to fundamental properties based on molecular dynamics and statistics such as linear rheological parameters and mechanical relaxation behavior. These properties for tree-shaped molecules can be characterized not by the adjacency matrix of their ordinal graph  $G$  but by that of their line graph  $L(G)$ . Consequently, the condition of determining whether the skeletal structure of highly branched chains can be uniquely determined from the relaxation

<sup>2</sup> Two graphs are isomorphic if there exists between their vertex sets a one-to-one correspondence having the property that, whenever two vertices are adjacent in either graph, the corresponding two vertices are adjacent in the other graph.

spectrum or not can be examined not by the existence of the isospectral graphs but by the existence of the isospectral line graphs.

**Appendix A**

Before going to the main subject, we prove the following relationship:

$$\Phi(\mathbf{R}_N; \lambda) = (2 - \lambda)\Phi(\mathbf{R}_{N-1}; \lambda) - \Phi(\mathbf{R}_{N-2}; \lambda), \tag{A.1}$$

where  $\mathbf{R}_N$  is the Rouse matrix of a tree-like chain with  $N$  vertices. Any tree digraph  $D_N$  of order  $N$  is constructed from one vertex being connected with a vertex in  $D_{N-1}$  by a directed edge (see figure 2). Before each step, the vertices and edges are renumbered, if necessary, so that the directed edge to be connected has its origin at the highest-numbered vertex. This ensures that the last row and the last column of  $\mathbf{R}_N$  have an element  $-1$  in their penultimate,  $+2$  in the last, and empty in the other positions as follows:

$$\mathbf{R}_N = \begin{bmatrix} \boxed{\mathbf{R}_{N-1}} & \begin{matrix} 0 \\ \vdots \\ 0 \end{matrix} \\ \begin{matrix} 0 \cdots \cdots 0 \end{matrix} & \begin{matrix} -1 \\ 2 \end{matrix} \end{bmatrix}. \tag{A.2}$$

The renumbering process is independent of the characteristic polynomial because it merely induces the same permutation in both the row-set and column-set of the determinant. Expanding  $\text{Det}|\mathbf{R}_N - \lambda\mathbf{E}_N|$  by its last column and row, we obtain

$$\text{Det}|\mathbf{R}_N - \lambda\mathbf{E}_N| = (2 - \lambda)\text{Det}|\mathbf{R}_{N-1} - \lambda\mathbf{E}_{N-1}| - (-1)^2 \text{Det}|\mathbf{R}_{N-2} - \lambda\mathbf{E}_{N-2}|. \tag{A.3}$$

This indicates that equation (A.1) is true for all positive integers  $N$ . Also, setting  $\lambda = 0$  in equation (A.3), we have  $\text{Det}|\mathbf{R}_N| = N$  [14].

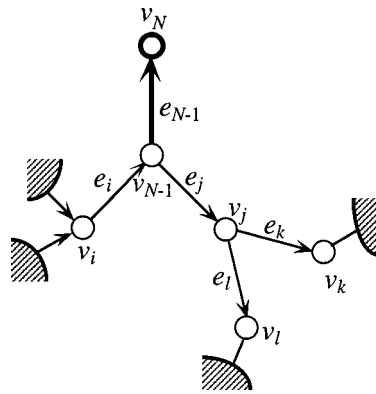


Figure 2. A tree digraph constructed by  $N$ th stepwise.



Using the same type of arguments, it can be shown that the matrix  $\mathbf{K}_N$  can be written as

$$\mathbf{K}_N = \begin{bmatrix} & & & & 0 \\ & & & & \vdots \\ & & & & 0 \\ & & \boxed{\mathbf{K}_{N-1}} & & 1 \\ 0 \cdots \cdots 0 & & & 1 & 2 \end{bmatrix}. \quad (\text{A.4})$$

Similarly, we have

$$\Phi(\mathbf{K}_N; \lambda) = (2 - \lambda)\Phi(\mathbf{K}_{N-1}; \lambda) - \Phi(\mathbf{K}_{N-2}; \lambda). \quad (\text{A.5})$$

Let us return to the proof of the following proposition:

$$\Phi(\mathbf{R}_N; \lambda) = \Phi(\mathbf{K}_N; \lambda). \quad (\text{A.6})$$

The proposition is certainly true for tree-like graphs containing  $N = 1, 2, 3, 4, 5$ , i.e.,  $\Phi(\mathbf{R}_1; \lambda) = \Phi(\mathbf{K}_1; \lambda)$ ,  $\Phi(\mathbf{R}_2; \lambda) = \Phi(\mathbf{K}_2; \lambda)$ ,  $\Phi(\mathbf{R}_3; \lambda) = \Phi(\mathbf{K}_3; \lambda)$ ,  $\Phi(\mathbf{R}_4; \lambda) = \Phi(\mathbf{K}_4; \lambda)$ ,  $\Phi(\mathbf{R}_5; \lambda) = \Phi(\mathbf{K}_5; \lambda)$ . Since we have equations (A.1) and (A.5), we can conclude by induction that equation (A.6) is true for all positive integers  $N$ .

## Appendix B

Let  $Z$  be the configuration partition function. The distribution function for  $S^2$  can be written as

$$P(S^2) dS^2 = \frac{dS^2}{Z} \int \delta(S^2 - N^{-1} \text{Tr}(\mathbf{r}\mathbf{r}^T)) e^{-(d/2b^2) \text{Tr}(\mathbf{r}\mathbf{Z}\mathbf{r}^T)} d\mathbf{r}, \quad (\text{B.1})$$

where

$$Z = \int e^{-(d/2b^2) \text{Tr}(\mathbf{r}\mathbf{Z}\mathbf{r}^T)} d\mathbf{r}.$$

The Fourier representation of the delta function gives [11]

$$P(S^2) dS^2 = \frac{dS^2}{2\pi} \int_{-\infty}^{\infty} \rho(ik) e^{ikS^2} dk \quad (\text{B.2})$$

and

$$\rho(ik) = Z^{-1} \int e^{-(d/2b^2) \text{Tr}[\mathbf{r}(\mathbf{Z} + ik(2b^2/dN)\mathbf{E})\mathbf{r}^T]} d\mathbf{r}, \quad (\text{B.3})$$

where  $i = \sqrt{-1}$ . The  $\rho(ik)$  becomes the Fourier transform of  $P(S^2)$ , i.e., the characteristic function so that we have

$$P(S^2) = \mathcal{F}^{-1}[\rho(ik)]. \quad (\text{B.4})$$

Considering the transformation to normal coordinates, the integration in equation (B.3) can be solved by making the use of equation (3) in the text; we obtain

$$\rho(ik) = \text{Det} \left( \mathbf{E} + ik \frac{2b^2}{d} \mathbf{R}^{-1} \right)^{-d/2} \equiv \varphi(ik)^{-d/2}. \quad (\text{B.5})$$

Using the change of variable  $z = ik$ , equation (B.4) can be transformed to

$$P(S^2) = \mathcal{L}^{-1}[\rho(z)] = \mathcal{L}^{-1}[\varphi(z)^{-d/2}]. \quad (\text{B.6})$$

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