our conclusion that the ordered structure of PHP in solution is form B, with the γ-hydroxyl group forming an intrachain hydrogen bond with a backbone carbonyl oxygen. The possibility of such a hydrogen-bond formation was suggested earlier for the polymer $(Gly\text{-}Hyp)_n$ by Mattice and Mandelkern,[20] while Torchia[7] from proton NMR studies had proposed a similar scheme for PHP.

From theoretical considerations, it has been shown in the previous paper that, though the presence of the γ-hydroxyl groups does not put any stereochemical restriction on the PHP chain taking up a structure with all-cis units, no intrachain hydrogen bonds, either direct or water-bridged, are possible for this structure. Hence the absence of this structure may be due to the fact that, in solution, once three consecutive units take up trans orientations, a hydrogen bond is formed linking the γ-hydroxyl group of the $(i + 1)$ th residue to the carbonyl oxygen of the $(i - 1)$th residue, and the polypeptide chain is locked in this thermodynamically favorable conformation. Thus, the occurrence of the intrachain hydrogen-bonded B form structure readily explains the absence of mutarotation and the great stability of PHP in solution.

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References and Notes

(1) M. Bansal, S. K. Brahmachari, and V. Sasisekharan, *Macromolecules*, preceding paper in this issue.
(2) V. Sasisekharan, *Acta Crystallogr.*, **12**, 903–909 (1959).
(3) V. Sasisekharan, *J. Polym. Sci.*, **47**, 391–396 (1960).
(4) J. Kurtz, A. Berger, and E. Katchalski in "Recent Advances in Gelatin and Glue Research", G. Stainsby, Ed., Pergamon Press, London, 1958, pp 131–135.
(5) I. Z. Steinberg, W. F. Harrington, A. Berger, M. Sela, and E. Katchalski, *J. Am. Chem. Soc.*, **82**, 5263–5279 (1960).
(6) W. L. Mattice and L. Mandelkern, *Macromolecules*, **3**, 199–201 (1970).
(7) D. A. Torchia, *Macromolecules*, **5**, 566–569 (1972).
(8) E. D. Kaufman, C. F. Nawrot, and R. H. Bull. *Arch. Biochem. Biophys.*, **117**, 93–97 (1966).
(9) D_2O was used instead of H_2O, in order to avoid overlap between the O–H band of H_2O and the band due to the γ-OH group of hydroxyproline.
(10) R. Mandel and G. Holzworth, *Biopolymers*, **12**, 655–674 (1973).
(11) E. S. Pysh, *Biopolymers*, **13**, 1563–1571 (1974).
(12) G. N. Ramachandran and V. Sasisekharan, *Adv. Protein Chem.*, **23**, 283–438 (1968).
(13) J. Mitra and C. Ramakrishnan, *Int. J. Pept. Protein Res.*, **9**, 27–48 (1977).
(14) J. L. Bensing and E. S. Pysh, *Biopolymers*, **10**, 2645–2648 (1971).
(15) D. S. Clark and W. L. Mattice, *Macromolecules*, **10**, 369–376 (1977).
(16) S. Tanaka and H. A. Scheraga, *Macromolecules*, **8**, 623–631 (1975).
(17) D. A. Torchia, *Macromolecules*, **4**, 440–442 (1971).
(18) D. A. Torchia and J. R. Lyerla, Jr., *Biopolymers*, **13**, 97–114 (1974).
(19) M. J. Deveney, A. G. Walton, and J. L. Koenig, *Biopolymers*, **10**, 615–630 (1971).
(20) W. L. Mattice and L. Mandelkern, *Biochemistry*, **10**, 1926–1933 (1971).

Fourfold Helical Structures for Polypeptides†

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ABSTRACT: Fourfold helical structures for polypeptides and their association in regular lattices with interchain hydrogen bonds were investigated by model building studies. These studies revealed that stereochemically satisfactory fourfold helical sturctures are possible for polyglycine, polyproline, and copolymers of glycine and proline with two and four units in the monomer. In these structures the unit height h for the backbone has been found to be restricted from 2.7 to 3.1 Å with four peptide units per turn of the helix. Energetically both fourfold and threefold helical structures are equally favorable.

It is well known that there are three basic types of polypeptide conformations, namely, the α helix,[1] the extended β form,[2] and the triple helix.[3] Each of these structures can aggregate to form a fibre, the first and the last by close packing of the approximately cylindrical protofibrils and in the β form by stacking of sheets.[4]

The α helix, the β form, and the triple helix represent single, double, and triple chain hydrogen bonded arrangements. The single chain α helix in which we have intrachain hydrogen bonds nearly parallel to the helix axis has been experimentally shown to be right handed. Each chain in the β form is a twofold helix and the chains are aggregated in the form of sheets with hydrogen bonds nearly perpendicular to the chain axis. In polyglycine II[5] and in poly(L-proline) II,[6] each chain is considered to be a threefold helix. In polyglycine II the chains are packed in a hexagonal array, each chain being hydrogen bonded to its six neighbors. These hydrogen bonds lie roughly perpendicular to the helix axis, and run in several directions. This type of triple chain arrangement with interchain hydrogen bond network throughout is possible only for glycyl residue. The three types of regular structures for polypeptides are shown in the (ϕ, ψ)-map in Figure 1. In this figure, the relevant parameters of a helix, n (number of residues per turn) and h (unit height), are also shown as contours, along with the observed (ϕ,ψ) values of glycyl and prolyl residues taken from available single-crystal data on linear peptides.[7] In addition to the above three types of structures it is known that the ω form of poly(β-benzyl L-aspartate) is a fourfold helix.[8] The ω helix like the α helix also has intrachain hydrogen bonds roughly parallel to the helix axis.

To the best of our knowledge there has been no report on fourfold helical structures and their association in a regular lattice with the interchain hydrogen bonding

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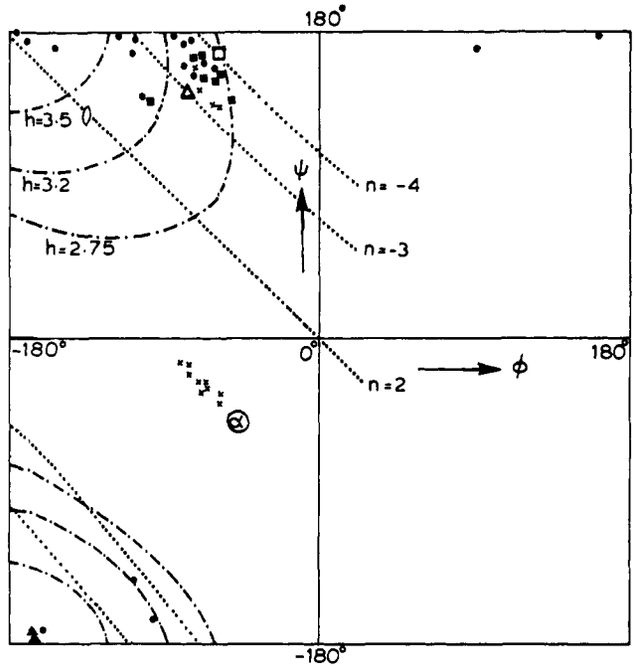

Figure 1. Standard structures such as α helix (α), β form (0), and triple helix (Δ) are shown in the (ϕ,ψ) map; a $\square$ denotes the fourfold structure. Contours ··· and --- denote helical parameters n and h, respectively. The conformations as observed in single crystals of linear peptides corresponding to residues Gly (●), Pro (×), Gly (▲) in sequence Gly-Gly and Pro (■) in sequence Pro-Pro are plotted.

scheme. Our model building investigations revealed that it is possible to build stereochemically satisfactory fourfold helical structures for polyglycine, poly(proline), and copolymers of glycine and proline. We have investigated regular fourfold helical structures with a network of interchain hydrogen bonding schemes in a lattice. We found that such a network of hydrogen bonding arrangement was possible only for polyglycine as no other side chain could be accommodated in the lattice. In the case of imino acid residues, proline and hydroxyproline, which are devoid of NH···O hydrogen bonds, regular fourfold helical structures were considered from packing considerations only. For copolymers of glycine and imino acid residues regular fourfold helical structures for the backbone with the formation of all the possible NH···O hydrogen bonds in a regular lattice were considered.

Homopolypeptides

(a) Polyglycine. For polyglycine regular fourfold helical structures with interchain hydrogen bonds for both parallel and antiparallel arrangements are possible depending upon the unit height h and the criteria applied for the hydrogen bond formation. The unit height h is found to be restricted from 2.7 to 3.1 Å for a hydrogen bond length (N···O) of 2.8 to 3.0 Å and an angle HNO <30°, both within the limits for NH···O hydrogen bond formation.[9] We have shown as an example one typical fourfold structure, projection down the helix and a side view, in Figures 2a and 2b corresponding to $n = -4$ and $h = 2.8$ Å. The hydrogen bond parameters for this structure are N···O = 2.9 Å and ∠HNO = 14°. This fourfold structure is shown in Figure 1 by an open square. The (ϕ,ψ) values for this structure are (-59°,168°) and the corresponding values for the threefold structure are (-76°,144°). It will be noticed that the difference between a threefold structure and a fourfold structure for a particular h lies essentially in the ψ value. In the fourfold type of arrangement, the chains are packed in a tetragonal unit

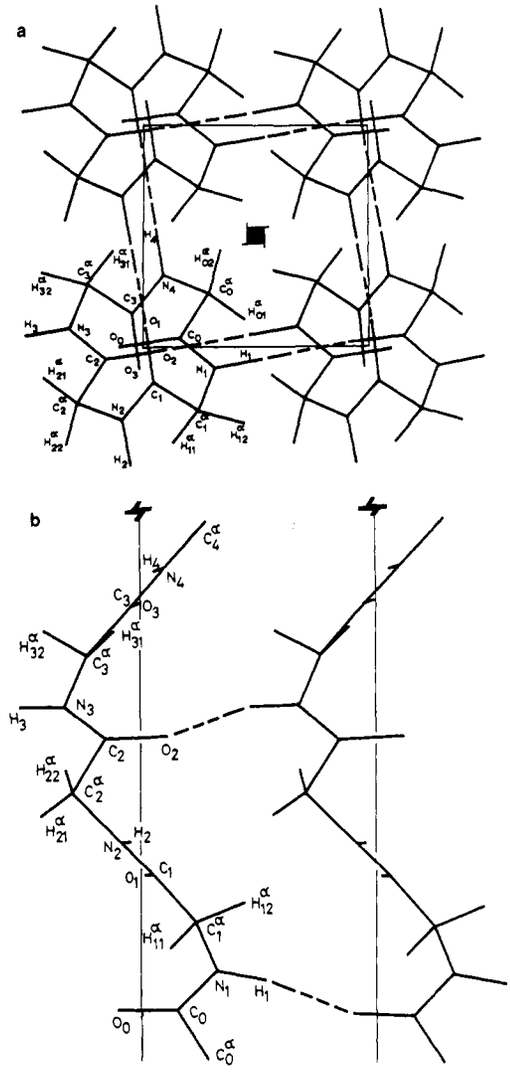

Figure 2. (a) Projection down the helix axis of a fourfold structure for polyglycine. Dashed lines denote hydrogen bonds. Note that the hydrogen bonds run almost in perpendicular directions linking neighboring chains. (b) A side view of the fourfold polyglycine structure. Note that hydrogen bonds (dashed lines) are roughly perpendicular to the chain axis.

cell, each chain being hydrogen bonded to its four nearest neighbors, consistent with the fourfold symmetry. The hydrogen bonds lie nearly perpendicular to the chain axis and run in perpendicular directions. For the arrangement cited here, the unit cell parameters are $a = b = 4.75$ Å and $c = 11.2$ Å and the space group is $P4_3$. It may be noted that both left- and right-handed structures are equally possible for polyglycine. In other words the space group can be $P4_1$.

In the literature polyglycine has been characterized so far in two forms, polyglycine I (PG I) and polyglycine II (PG II). Both forms have been studied by powder X-ray diffraction,[10,11,5] electron diffraction,[12,13] infrared,[14] and theoretical methods.[15,16] Form I has been characterized as extended β form.[12] Although the X-ray powder pattern of PG II shows five rings, only two rings have been used for characterizing the structure.[5] No further details of spacings or relative intensities were given. However, the data have been interpreted in terms of a threefold structure packed in a hexagonal array.[5] The paucity of X-ray data do not permit a detailed investigation of the structure. An attempt must be made to get more diffraction data on different forms of polyglycine to arrive at their structures.

However, we have analyzed single-crystal data of linear peptides to see the types of regular helical structures that can be generated for polyglycine from the observed conformational angles of glycyl residues.[7] The (ϕ,ψ) values of glycyl residues in linear peptides are plotted in Figure 1. It will be noticed that whereas the ϕ values are scattered the ψ values are clustered near the 180° region. Interestingly, not a single conformation is observed in the α-helical region. There is only one linear peptide having the sequence Gly-Gly for which single-crystal data are available. The ϕ,ψ values for this lie near the n = 2 contour and also to the rippled sheet structure proposed for PG I by Lotz.[12] A significant number of glycyl residues are clustered in the region enclosed by n = −3, n = −4, and h = 2.75 and 3.2 Å contours. In this region the ψ values are centered above and below the typical fourfold structure but only above the threefold structure. Interestingly, the average ψ value for all the glycyl residues is near to the fourfold structure. We have also computed the packing ratio P given by ($\rho(\triangle)/\rho(\square)$), where $\rho(\triangle)$ and $\rho(\square)$ are theoretical densities of the typical threefold and fourfold structures, respectively. This ratio turned out to be approximately unity when the hydrogen bond length in both the structures was the same. Also the single chain conformational energies for the two types of structures are found to be nearly the same (refer to Figure 3 in ref 17, where it will be noticed that there is a flat minimum in the region of threefold and fourfold structures). Thus, our studies strongly suggest the possibility of fourfold regular helical structure for polyglycine with a interchain hydrogen bond network in a lattice. The observed ψ values can also lead to the generation of nonintegral helices for polyglycine. However, such nonintegral helices cannot be packed in a regular lattice with all NH and CO groups directly participating in hydrogen bonds.

(b) Poly(L-proline). For poly(L-proline) also regular fourfold helical structures are possible with unit height h ranging from 2.7 to 3.0 Å. Conformational energy calculations on the ring puckering of the pyrrolidine ring have indicated that ϕ can vary from −35 to −85°.[18] This is precisely the range of ϕ that is possible for threefold and fourfold helical structures.

Poly(L-proline) has so far been characterized in two conformational forms as studied by X-ray diffraction,[19-22,6] ORD–CD,[23] and theoretical analysis.[24-27] Form I is a right-hand helix with all cis residues containing 3.33 residues/turn and h = 1.9 Å. Form II is a left-hand helix of trans residues with n = −3 and h = 3.12 Å. However, different modifications of poly(L-proline) have been observed when treated with different solvents and precipitated under various conditions.[20] These modifications have neither been studied in detail nor their structures reported.

As before, we have plotted in Figure 1 the conformational angles of prolyl residues as observed in linear peptides to see the types of helical structures that can be generated using these values. It will be noticed that the (ϕ,ψ) values are clustered in two regions with ϕ = −80 to −45° and ψ = −40 to −15° in one case and ψ from 135 to 170° in the other case for the same values of ϕ. The pyrrolidine–pyrrolidine sequence in linear peptides, however, lies near $\psi \approx 160°$ between n = −3 and n = −4 contours suggesting regular threefold and fourfold helical structures as well as nonintegral helices for polyproline. In order to see the relative stabilites of threefold and fourfold helical structures we have considered the packing ratio and energetics of these structures. The calculated packing ratio in the case of threefold poly-L-proline II with the fourfold poly(L-proline) (with chain-to-chain separation of about 8.5 Å and h = 2.8 Å) is again approximately unity. Also the energy difference between fourfold structure and the threefold structure is negligible.[27] However, as stated earlier the X-ray data on poly(L-proline) II are not in confirmity with the fourfold helical structure. Therefore, evidence for the fourfold helical structure should come from X-ray studies and we are looking into the various powder patterns obtained for poly(L-proline) when treated with different solvents and precipitated under various conditions.[20]

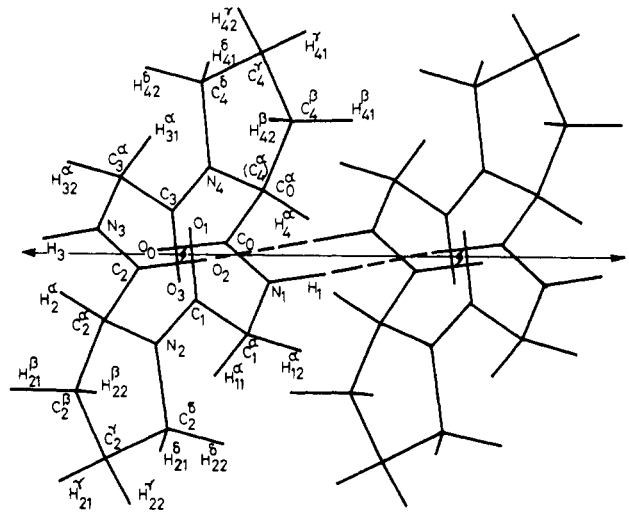

Figure 3. Projection down the helix axis of two chains of poly(Gly-L-Pro) with a fourfold structure for the backbone. Note that the hydrogen bonds are confined to a sheet along the arrow shown, similar to the β form. Extension of the structure in space will lead to roughly parallel hydrogen bonded networks, again as in the β structure. Packing between successive chains is achieved by close packing of the pyrrolidine rings.

Copolymers of Glycine and Proline

An extension of the above ideas indicates the possibility of fourfold helical structues for the backbone of copolymers containing glycyl and prolyl residues. We have carried out model building studies for polymers containing two, three, and four residues in the monomer unit. In all our studies we have considered only trans peptide structure for Gly-L-Pro following the crystal structure data of linear peptides, which indicate trans as the most favored conformation. The occurrence of *trans*-Gly-L-Pro has also been verified by NMR.[28]

Polydipeptides

Poly(Gly-L-Pro) and Poly(Gly-L-Hyp). Circular dichroism[29] and NMR[30,31] studies have been carried out on poly(Gly-L-Pro). The solution studies have been interpreted in terms of an unordered structure for this copolymer[29] and in terms of cis–trans isomerization of the peptide bond.[30] Also isomerization about the C^{α}–C bond in the L-prolyl residue has been suggested.[32] On the other hand, the solution studies of poly(Gly-L-Hyp) have been interpreted as due to an ordered structure of this copolymer.[29] However, no satisfactory structure has been proposed for this copolymer. The standard structures like α helix, β form, and triple helix are not possible for both copolymers. Therefore, we present an ordered structure for poly(Gly-L-Pro), in which Gly and Pro alternate within a fourfold structure for the backbone of the polypeptide. A structure is shown in Figure 3, which is a projection down the helix axis with h = 2.8 Å. So far, there has been no report of X-ray diffraction studies on these copolymers to arrive at definitive conclusions with regard to their structures.

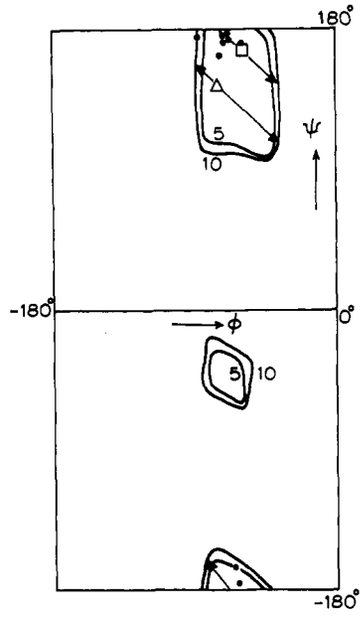

Figure 4. The observed conformation ψ_1 of Gly and ϕ_2 of Pro in the sequence Gly-Pro as observed in single crystal structures of linear peptides are plotted (denoted by ●) in the (ϕ,ψ) plane. The allowed ranges for the fourfold and threefold structures are denoted by (←□→) and (←△→). Energy contours for poly(L-proline) (—) corresponding to 5 and 10 kcal/mol are shown.

However, an indication to the possible structures of poly(Gly-L-Pro) can be obtained from the crystal data involving sequence Gly-L-Pro. Analysis of the crystal data containing the Gly-L-Pro sequence in linear peptides again indicated that $\psi_{(Gly)}$ is restricted to 180 ± 15° only. The ϕ value of proline, as is well known, is restricted to −60 ± 20°. To emphasize this observation, we have plotted in Figure 4 the ψ value of glycine and the ϕ value of proline as observed in crystal structures of linear peptides involving the sequence Gly-L-Pro. In this figure also are marked the allowed ranges of regular fourfold and threefold structures (or helices) consistent with the hydrogen bond criteria. These are precisely the ranges possible (within 5 and 10 kcal/mol energy contours for poly(L-proline)) for regular helical structures of copolymers containing prolyl residues and having the same (ϕ,ψ) values throughout the backbone. It will be noticed that the observed values cluster only around the contour $n = -4$, strongly suggesting the possibility of fourfold structure for poly(Gly-L-Pro).

For poly(Gly-L-Hyp) in addition we find that the hydroxyl groups of the hydroxyproline can participate in intersheet hydrogen bond interactions leading to additional stability. The detailed analysis is in progress and this will be reported elsewhere.

Polytripeptides

Regular fourfold structures for the backbone are not possible for the polytripeptides containing glycine and proline residues, as a network of interchain hydrogen bonds between NH and carboxyl groups cannot be formed.

Polytetrapeptides

Ordered structures of the fourfold type can be built for polytetrapeptides containing glycine and proline.

Poly(Gly-Gly-Gly-X), X = Pro or Hyp. For poly(Gly-Gly-Gly-X), with X = L-Pro or L-Hyp, fourfold structures can be built with all the available NH groups participating in hydrogen bonds. As an example, for poly(Gly-Gly-Gyl-L-Pro), corresponding to $n = -4$ and $h = 2.8$ Å for the backbone, the projection down the helix is given in Figure 5. In fact, poly(Gly-Gly-L-Pro-Gly) and poly(Gly-Gly-L-Hyp-Gly) (a different designation of poly(Gly-Gly-Gly-L-Pro) and poly(Gly-Gly-Gly-L-Hyp), respectively) have been studied in solution.[30] Under the conditions studied, the latter is said to be in an ordered conformation while the former is not ordered. It has also been pointed out that it was difficult to propose an ordered structure for poly(Gly-Gly-L-Hyp-Gly). A triple helical structure for this copolymer was attempted and was found to be inconsistent with the experimental data and therefore a definitive conclusion in regard to the actual structure was not made.[30] However, we find that regular fourfold structures for the backbone of the polypeptide with hydrogen bond network can be built, as shown in Figure 5.

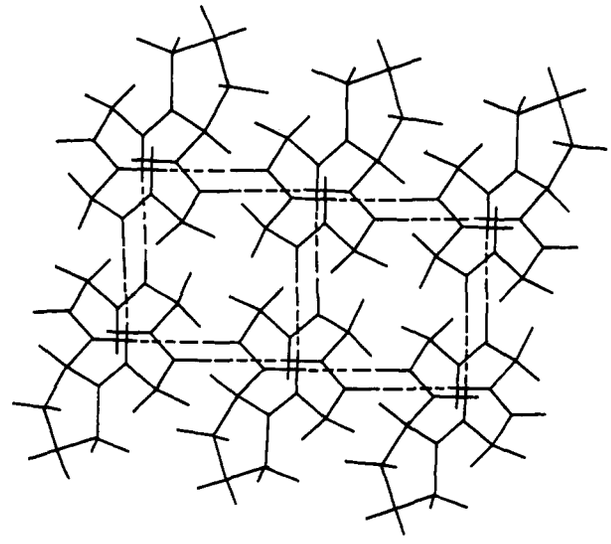

Figure 5. Projection down the helix axis of poly(Gly-Gly-Gly-L-Pro) with a fourfold structure for the backbone.

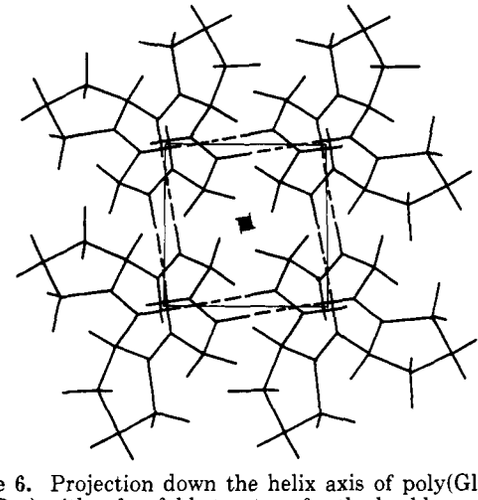

Figure 6. Projection down the helix axis of poly(Gly-Gly-L-Pro-L-Pro) with a fourfold structure for the backbone showing tetrameric association of chains.

Poly(Gly-Gly-X-Y), X = Pro or Hyp and Y = Pro or Hyp. For the copolymer of the type poly(Gly-Gly-X-Y) with X and Y = L-Pro or L-Hyp, satisfactory fourfold structure can be built, again with all the NH groups involved in interchain hydrogen bonds. A projection down the helix axis for poly(Gly-Gly-L-Pro-L-Pro) is illustrated in Figure 6 (corresponding to $n = -4$ and $h = 2.8$ Å for the backbone). Further, association of such tetrameric chains can lead to coiled coil structures similar to the triple helical structures. Work on these lines is in progress.

Fourfold structures of these types can also be built for other sequential tetrapeptides with glycyl and prolyl or

hydroxyprolyl residues. The details of these will be published elsewhere.

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References and Notes

(1) L. Pauling, R. B. Corey, and H. R. Branson, *Proc. Natl. Acad. Sci. U.S.A.*, **37**, 205 (1951).
(2) L. Pauling and R. B. Corey, *Proc. Natl. Acad. Sci. U.S.A.*, **39**, 253 (1953).
(3) G. N. Ramachandran and G. Kartha, *Nature (London)*, **176**, 593 (1955).
(4) G. N. Ramachandran and V. Sasisekharan, *Adv. Protein Chem.*, **23**, 283 (1968).
(5) F. H. C. Crick and A. Rich, *Nature (London)*, **176**, 780 (1955).
(6) V. Sasisekharan, *Acta Crystallogr.*, **12**, 897 (1959).
(7) E. Bendetti in "Proceedings of the Fifth American Peptide Symposium", M. Goodman and J. Meienhofer, Eds., Wiley, New York, N.Y., 1977, p 257.
(8) E. M. Bradbury, L. Brown, A. R. Downie, A. Elliott, R. D. B. Fraser, and W. E. Hanby, *J. Mol. Biol.*, **5**, 230 (1962).
(9) C. Ramakrishnan and N. Prasad, *Int. J. Protein Res.*, **3**, 209 (1971).
(10) C. H. Bamford, L. Brown, A. Elliott, W. E. Hanby, and I. F. Trotter, *Nature (London)*, **171**, 1149 (1953).
(11) C. H. Bamford, L. Brown, E. M. Cant, A. Elliott, W. E. Hanby, and B. R. Malcolm, *Nature (London)*, **176**, 396 (1955).
(12) B. Lotz, *J. Mol. Biol.*, **87**, 169 (1974).
(13) F. J. Padden and H. D. Keith, *J. Appl. Phys.*, **36**, 2987 (1965).
(14) T. Miyazawa in "Poly-α-Amino Acids", G. D. Fasman, Ed., Marcel Dekker, New York, N.Y., 1967, Chapter 2.
(15) F. Colonna-Cesari, S. Premilat, and B. Lotz, *J. Mol. Biol.*, **87**, 181 (1974).
(16) G. N. Ramachandran, C. Ramakrishnan, and C. M. Venkatachalam in "Conformation of Biopolymers", Vol. 2, G. N. Ramachandran, Ed., Academic Press, New York, N.Y., 1967, p 429.
(17) R. A. Scott and H. A. Scheraga, *J. Chem. Phys.*, **45**, 2091 (1966).
(18) C. M. Venkatachalam, B. J. Price, and S. Krimm, *Macromolecules*, **7**, 212 (1974).
(19) P. M. Cowan and S. McGavin, *Nature (London)*, **176**, 501 (1955).
(20) V. Sasisekharan, *J. Polym. Sci.*, **47**, 373 (1960).
(21) W. Traub and U. Shmueli in "Aspects of Protein Structure", G. N. Ramachandran, Ed., Academic Press, New York, N.Y., 1963, p 81.
(22) W. Traub and U. Shmueli, *Nature (London)*, **198**, 1165 (1963).
(23) I. Z. Steinberg, W. F. Harrington, A. Berger, M. Sela, and E. Katchalaski, *J. Am. Chem. Soc.*, **82**, 5263 (1960).
(24) P. De Santis, E. Giglio, A. M. Liquori, and A. Ripamonti, *Nature (London)*, **201**, 456 (1965).
(25) P. R. Schimmel and P. J. Flory, *Proc. Natl. Acad. Sci. U.S.A.*, **58**, 52 (1967).
(26) N. Gō and H. A. Scheraga, *Macromolecules*, **3**, 188 (1970).
(27) W. L. Mattice, K. Nishikawa, and T. Ooi, *Macromolecules*, **6**, 443 (1973).
(28) G. Schilling and Kricheldorf, *Makromol. Chem.*, **178**, 885 (1977).
(29) W. L. Mattice and L. Mandelkern, *Biochemistry*, **10**, 1926 (1971).
(30) D. A. Torchia, *Biochemistry*, **11**, 1462 (1972).
(31) D. A. Torchia and J. R. Lyerla, Jr., *Biopolymers*, **13**, 97 (1974).
(32) D. A. Rabenold, W. L. Mattice, and L. Mandelkern, *Macromolecules*, **7**, 43 (1974).

Transport Coefficients of Helical Wormlike Chains. 2. Translational Friction Coefficient

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ABSTRACT: The translational friction coefficient of the helical wormlike chain is evaluated by an application of the Oseen–Burgers procedure of hydrodynamics to the cylinder model. The Oseen hydrodynamic interaction tensor is preaveraged, and therefore there is no need of consideration of the effect of the coupling between translational and rotational motions. A useful empirical interpolation formula is also derived to be valid for any possible values of the model parameters. Some salient aspects of the behavior of the sedimentation coefficient are discussed on the basis of the numerical results. In particular, it is pointed out that even if the proportionality of the sedimentation coefficient to the square root of the molecular weight is experimentally observed over a wide range, the chain may not always be regarded as a random coil but may possibly be characterized as a helical wormlike chain.

In a previous paper,[1] part 1 of this series, a study of the steady-state transport coefficients of helical wormlike (HW) chains[2,3] (without excluded volume) was initiated. The evaluation of the translational diffusion (or friction) coefficient and intrinsic viscosity of the characteristic regular helix,[3] i.e., one of the three extreme forms of the model chain corresponding to its minimum configurational energy, was carried out by an application of the Oseen–Burgers procedure of hydrodynamics[4] to cylinder models. In this paper, we proceed to evaluate the translational friction coefficient of HW chains along the same line.

As discussed in detail previously,[1] there are two fundamental problems to be considered. One is concerned with the steady-state transport length scales for the cylinder model, and the other with the possible effect of the coupling between translational and rotational motions of skew bodies. The length scales to be adopted must be of the same order as or larger than those associated with the equilibrium chain configurations. This should always be kept in mind when an analysis of experimental data is made by the use of theoretical expressions for the transport coefficients derived in the present and later papers. As for the second problem, it was shown that if the Oseen hydrodynamic interaction tensor is preaveraged in the Kirkwood–Riseman scheme,[5] the coupling does not affect the translational friction coefficient of the regular helix at all, while the effect on its intrinsic viscosity may be negligibly small except when the number of its turns is very small. It is clear that the skewness is partly destroyed for the HW chain having internal degrees of freedom, and moreover, the evaluation for this chain is carried out with the preaveraged Oseen tensor as in the case of the Kratky–Porod (KP) wormlike chain.[4] Therefore, we do not consider the coupling in the present and later papers.

The mean reciprocal distance between two contour points of the chain, which is necessary for the evaluation

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