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Life Before RNA

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ABSTRACT

The hypothesis that life originated and evolved from linear informational molecules capable of facilitating their own catalytic replication is deeply entrenched. However, widespread acceptance of this paradigm seems oblivious to a lack of direct experimental support. Here, we outline the fundamental objections to the *de novo* appearance of linear, self-replicating polymers and examine an alternative hypothesis of template-directed coding of peptide catalysts by adsorbed purine bases. The bases (which encode biological information in modern nucleic acids) spontaneously self-organize into two-dimensional molecular solids adsorbed to the uncharged surfaces of crystalline minerals; their molecular arrangement is specified by hydrogen bonding rules between adjacent molecules and can possess the aperiodic complexity to encode putative protobiological information. The persistence of such information through self-reproduction, together with the capacity of adsorbed bases to exhibit enantiomorphism and effect amino acid discrimination, would seem to provide the necessary machinery for a primitive genetic coding mechanism. Key Words: RNA—Life—Information—Self-replication—Self-organization. Astrobiology 2, 231–239.

INTRODUCTION

ELUCIDATION OF ITS DOUBLE-HELICAL STRUCTURE supported the identification of DNA as the postulated "aperiodic solid" of information-bearing chromosomes (Schrödinger, 1944) and provided a mechanism for the molecular basis of heredity (Watson and Crick, 1953a,b). Information in nucleic acids is conveyed by a linear, irregular sequence of nucleotides that encodes the construction of protein catalysts essential for life. The nucleotides specify the linear sequences of L-α-amino acids (Crick, 1958), whose linear arrangement is the primary determinant of protein structure and function (Anfinson, 1973). The peptide bond, the chemical linkage between amino acid residues, is formed in a dehydration reaction between the ammonium and carboxylate moieties of adjacent amino acid molecules. The resulting polypeptide chains fold into complex three-dimensional solids with the capacity to catalyze specific chemical reactions.

In modern cells, the transfer of information from DNA to protein is mediated by RNA molecules. However, in addition to encoding protein sequences, RNA also plays a catalytic role in the formation of the peptide bond (Khaitovich *et al.*, 1999). Other naked RNA sequences exhibit limited catalytic activity on a variety of cellular substrates (Yarus, 1999), and, in the laboratory, RNA catalysis has been engineered by rounds of *in vitro* mutation, amplification, and selection—a process of forced Darwinian evolution (Beaudry and Joyce, 1992). Such techniques could ultimately yield artificial self-replicating polymers (Johnston *et al.*, 2001). The capacity for RNA sequences to convey information (genotype), catalyze chemical reactions (phenotype), and undergo Darwin-

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ian evolution lends support to the idea that the very first forms of chemical "life" were naked RNA molecules capable of facilitating their own replication and evolution (Gilbert, 1986).

The prebiotic appearance of the purine coding bases of RNA, adenine and guanine, seems straightforward. Simulation experiments of the chemistry of the primitive Earth (Miller, 1987) and the atmosphere of the Jovian moon Europa (Levy *et al.,* 2000) and analyses of organic material recovered from carbonaceous chondrites (Cronin, 1998) yield various mixtures of amino acids and some of the purine bases of RNA. These products are strongly indicative of hydrogen cyanide–based chemistry as the *first* source of primordial organic species (Ferris and Hagan, 1984; Ferris, 1992; Minard *et al.,* 1998).

The chemical problems with the *de novo* prebiotic appearance of RNA center predominantly on difficulties with the prebiotic origin of the sugar–phosphate scaffold (Schwartz and de Graaf, 1993) that forms the pseudocrystalline backbone of the linear polymer. A prebiotic route to the sugar is not trivial because carbohydrates are born of chemistry incompatible with the cyanide route to the bases (Arrhenius *et al.,* 1994). At the same time, the laboratory synthesis of linear RNA polymers on the surfaces of clay catalysts from artificially activated mononucleotide precursors (Ferris and Ertem, 1992) obscures the real-world problems of premature chain termination (Shapiro, 2000) and enantiomeric cross-inhibition (Joyce *et al.,* 1987). This has led to the suggestion that RNA was preceded by informational polymers with simpler chemical backbones that did not contain the ribose sugar moieties but still possessed the functional coding elements, the purine and pyrimidine bases (Joyce *et al.,* 1987; Nelson *et al.,* 2000). This scenario is appealing because of the way that nucleic acids transmit their genetic information from one generation to the next. The hydrogen bond pairing of complementary bases provides a mechanism for a linear, connected, evolutionary hierarchy of polymer sequences.

For the polymer hypotheses, the chemical difficulties of polymerization are further complicated by the susceptibility of the RNA bases to aqueous-phase hydrolysis (Shapiro, 1995, 1999; Levy and Miller, 1998), a process accelerated at elevated temperatures (Levy and Miller, 1998). Prevailing conditions could have alleviated this problem, since the bases are known to adsorb spontaneously to the surfaces of uncharged mineral species (Winter and Zubay, 1995; Cohn *et al.,* 2001; Sowerby *et al.,* 2001a,c). Such adsorption could have protected them from hydrolytic attack within the prebiotic environment, but would also have removed them from the aqueous phase and prevented them from participating in chemical processes leading to polymers or their monomeric building blocks.

Quite apart from their fundamental chemical difficulties, the origin of informational, self-replicating polymers also suffers from an informational paradox—the origin of the first such molecules (Cairns-Smith, 1982; Yockey, 1992; Shapiro, 2000). The odds are stacked against the random, *de novo* appearance of exceptionally rare polymer sequences, and that begs for a deterministic rather than stochastic origin. Such considerations inevitably force us to consider the possibility that life arose from simpler processes that did not rely on the primary involvement of linear, self-replicating molecules.

GENETIC TAKEOVER

Chemical and cellular life can be regarded as distinct because the properties of living systems are not necessarily limited to those systems encapsulated by a cellular membrane (Langton, 1989) and a certain level of chemical evolution may have occurred before the development of a cellular apparatus (Oparin, 1957). The "double-origin" hypothesis of Dyson (1985) envisages the spontaneous origin of a protein-catalyzed metabolism capable of self-reproduction. By-products of the metabolism enabled a *second* origin, giving rise to replicating RNA that parasitized the metabolism. This scenario is chemically less demanding than a *de novo* RNA world because the problems with RNA synthesis and replication can be overcome by utilizing the machinery of the metabolism. Dyson likened this to a bacteriophage utilizing the host biochemistry of a bacterium to complete the replication part of its life cycle. The difference between *self-reproduction* and *self-replication* is made distinct. Self-replication implies direct copying from a parent structure. Self-reproduction, on the other hand, does not require heredity in the first instance, but such a system can nevertheless seed subsequent generations.

Cairns-Smith (1982) argued for a previtral, naked genetic element, G_1, that somehow influenced its

environment to ensure its perpetuation. The magnitude of such an influence would not be important, but any manifestation of environmental change would, in essence, be the phenotype of G_1, which, in addition to tipping the scales in favor of perpetuating G_1, would provide an environment conducive to organic chemistry. Cairns-Smith (1982) suggested that the adsorptive and catalytic properties of clays could have been the instruments of chemical change and that the aperiodic arrangement of defect structure in their crystalline solid phase could have provided the architecture for replicative information processing.

Although their scenarios are superficially different, Dyson (1985) and Cairns-Smith (1982) concur: A prenucleic acid organic chemistry on the primitive Earth provided the metabolic architecture of catalysts for the subsequent synthesis of informational nucleic acids, which may have originally played a role in catalysis and, also, have been used for purposes other than the storage of genetic information. The distinction between these two scenarios is that, in Dyson's proposition, the formation of such a catalytic metabolism was spontaneous—it was *not* preceded by an informational architecture, as required by Cairns-Smith's proposal.

It is technically challenging to see how Dyson's "metabolism" dependent upon protein catalysts could arise spontaneously and then evolve independently of an informational architecture. Nonbiological amino acid polymerization can be achieved by dehydration on the surfaces of inorganic solids (Brack, 1993; Lahav, 1994; Rode, 1999), and the selective adsorption of chiral amino acid enantiomers has been demonstrated on chiral inorganic crystal faces (Hazen *et al.*, 2001). However, abiotic peptides have not generally been favored as prebiotic catalysts because noncoded polymerizations have to overcome astronomical odds to generate the specific amino acid sequences of biological proteins (Yockey, 1992). Furthermore, the formation of homochiral catalysts would require a symmetry break (Avetisov and Goldanskii, 1993), and the presence of interfering organic compounds (such as carboxylic acids and amines) would cause premature chain termination (Shapiro, 2000).

According to Cairns-Smith (1982), prevital genetic information *must* have been a prerequisite for evolution because *only* information can persist and *only* information can evolve. In his view, modern nucleic acids were established within the framework of a G_1-encoded "metabolism" and gradually developed the ability for coding and translation until they dominated the nature of the metabolism: G_1 was usurped and eventually replaced by the nucleic acid genetic elements (G_2). This was the essence of "genetic takeover."

Genetic takeover is a process that might have involved a number of different genetic materials. No linear connected hierarchy of genetic architectures needs to have existed, and subsequent genetic architectures would not necessarily bear any resemblance to their predecessors. In this situation, it would be impossible to deal with the technicalities of the origin of RNA in the absence of the chemical platform from which it *directly* originated.

If such a scenario is to be contemplated, the first problem, it seems, would be to demonstrate the feasibility of some persistent, non-nucleic acid, G_1, that has the ability to store information, evolve, confer phenotypic advantage on itself, and, at the same time, specify complex organic chemistry. Only then would the chemical architecture become sufficiently complex to lead to secondary genetic system(s) (G_2) from which nucleic acids could ultimately be derived.

NONPOLYMER INFORMATIONAL CHEMISTRY

Living systems exist in the solid regime in the vicinity of an order–chaos phase boundary (Kauffman, 1993). Nonlinear dynamic systems demonstrate complex behavior, adaptability, and computation in this transition region. Biochemically, such a boundary exists between the chaotic chemical milieu of the cell and the ordered solid of cellular structures. Molecules held rigid by covalent and noncovalent interactions form a solid phase. Free-moving particles distributed within the aqueous matrix are in the liquid phase. These definitions are, however, blurred at the cellular level. Cytoplasmic molecules are clearly dissolved, but the positions of their atomic building blocks are fixed relative to one another, and so they represent the germ of a solid phase (Schrödinger, 1944). By this reasoning, one might regard a complex protein molecule as a solid, whereas its molecular building block precursors, the amino acids, are able to move freely with respect to one another and are fluid. This subtle differ-

ence means that individual molecular species such as amino acids can serve as diffusible intermediates in chemical metabolism but become chemically redefined within the context of macromolecules.

During nucleic acid replication and protein synthesis, a phase transition occurs as chaotic-phase monomers are selectively solidified into new polymeric solids. Such processes all but eliminate the stochastic nature of chemical reactions because, at nucleic acid and protein surfaces, liquid-phase molecules cross a fluid–solid phase boundary and lose rotational and translational degrees of freedom. Upon adsorption, kinetic pathways are made available to otherwise unfavorable chemical reactions by facilitating appropriate collisions between surface-adsorbed species. If life is poised at the edge of chaos, it seems reasonable to suggest that life also originated there (Kauffman, 1993). If life is maintained and perpetuated at a solid–liquid interface, it is logical to focus investigation of the origins of life at such a phase boundary.

In the geochemical environment of the early Earth, the mineral–water interface would have been the dominant order–chaos phase boundary. Minerals have long been invoked in models for the origin of life (Bernal, 1951) and can provide surfaces for physical adsorption (Sowerby *et al.*, 1996; Parsons *et al.*, 1998; Smith, 1998), discrimination of chiral enantiomers (Hazen *et al.*, 2001), and catalytic polymerization of amino acids (Brack, 1993; Lahav, 1994; Rode, 1999) and nucleotides (Ferris and Ertem, 1992). Minerals can set the redox state of geochemical environments (Shock, 1990) and provide the reducing energy for the construction of biological precursors such as amino acids (Hennet *et al.*, 1992) and the establishment of pH gradients (Russell and Hall, 1997). Precipitated mineral particles can also provide vesicles for encapsulation (Russell and Hall, 1997).

A THEORY BASED ON EXPERIMENT

Scanning tunneling microscopy (Binnig *et al.*, 1982) is a powerful tool that we have used to investigate putative protobiological structures formed from the bases of nucleic acids adsorbed to inorganic surfaces (Sowerby *et al.*, 1996, 1998b, 2000, 2001b; Sowerby and Petersen, 1997). Our results have led us to the conclusion that the fundamental interactions of bases with mineral solids are an inescapable consequence of their prebiotic appearance in the geochemistry of the early Earth. Furthermore, we have developed a plausible hypothesis that such fluid–mineral interactions had a critical role in the origin of life (Sowerby *et al.*, 1996, 2000, 2001b). In our elaboration of genetic takeover, the prevital genetic element, G_1, is composed of the bases of nucleic acids, which form aperiodic, tessellated patterns on the surfaces of crystals. The "genotype" resides in their information-bearing arrangements, which obey strict interaction rules and so are of deterministic origin; "phenotype" is manifested in the form of prebiotic proteins synthesized on this primitive coding template of adsorbed bases. It is our hypothesis that these *encoded protein systems* had the capacity to evolve and so ultimately provide the chemical platform from which modern nucleic acids and protein translation originated.

The idea that a complex, encoded pre-RNA chemistry existed on the prebiotic Earth is radical, but rests on solid experimental evidence:

1. Simulation of the chemistry of the early Earth and extraterrestrial environments reveals that purine and pyrimidine bases and amino acids are contemporary products of nonbiological cyanochemistry (Miller, 1987; Minard *et al.*, 1998; Levy *et al.*, 2000) and indicates a noncontrived, putative availability of the relevant compounds.
2. The affinity of the bases for the surfaces of uncharged inorganic solids (Winter and Zubay, 1995; Cohn *et al.*, 2001; Sowerby *et al.*, 2001a) withdraws the bases from solution so that they are protected from hydrolysis. While such adsorption means that the free bases could not have participated in any aqueous-phase process leading to RNA, we postulate that this phenomenon could have been a primary prerequisite for the construction of G_1. The bases are planar molecules that can tessellate a surface in two dimensions to give arrays that are stabilized by van der Waals interactions with the underlying surface and by hydrogen bonds formed between adjacent molecules, a configuration originally postulated on the basis of thermodynamic measurements made at the mercury–water interface (Saffarian *et al.*, 1987). The hydrogen bond interactions between the bases are drawn from a large reper-

toire of possibilities, including those of Watson–Crick pairing found in nucleic acids, and the monolayers are constructed using physicochemical and geometric rules governing local interactions (Sowerby *et al.,* 2000). Electrochemical measurements show that maximal adsorption of the bases occurs at the potential of zero charge (Vetterl, 1966). The proposed mechanism of monolayer formation relies on the spatial restriction of solvent at the uncharged interface, where hydrogen bonding interactions between the bases are favored. This is in contrast to the aqueous phase, where the bases are surrounded by water molecules and remain dissolved (de Levie and Wandlowski, 1994). The interaction of nucleic acid bases with clays (Winter and Zubay, 1995) and graphite (Sowerby *et al.,* 2001a) shows that purine bases are, quantitatively, significantly more readily adsorbed than the pyrimidines and that this adsorption shows a strong temperature dependence (Sowerby *et al.,* 2001c). Although not all of the surfaces studied in model experiments can be considered dominant prebiotic materials, uncharged mineral surfaces were present in prebiotic times and would have facilitated the accretion of organic matter. Relevant examples come from the mineral families, zeolites, feldspars, and silicas (Smith, 1998). Bulk adsorption studies show that the purine base, adenine, adsorbs to clays (Winter and Zubay, 1995), pyrite (FeS_2), quartz (SiO_2), pyrrhotite (FeS), magnetite (Fe_3O_4), and forsterite (Mg_2SiO_4) (Cohn *et al.,* 2001). The geochemical prevalence of these minerals in the Hadean ocean would have allowed them to act as a sink for prebiotic purines. Removal of the bases from the aqueous phase would confer protection from hydrolysis, as well as establishing a nonequilibrium thermodynamic framework for increased base synthesis.

3. The spacing of the proton acceptors in the base arrays is consistent with peptide bond dimensions. Molecular modeling indicates that amino acids can form hydrogen bonds with the proton acceptors in the monolayers and are in the correct spatial arrangement for subsequent peptide formation (Sowerby and Heckl, 1998).
4. Spontaneous formation of peptide bonds between amino acids is facilitated by evaporation on the surfaces of inorganic solids (Brack, 1993; Lahav, 1994; Rode, 1999) under plausible prebiotic conditions.
5. Prebiotic chemistry, it seems, will have produced mixtures of both left- and right-handed versions of the amino acids; a symmetry break is required to separate one structural isomer from the other (Avetisov and Goldanskii, 1993). On certain minerals, the monolayers are enantiomorphic even though they are composed of achiral molecules (Sowerby *et al.,* 1996). The resulting symmetry break could be related to the peculiar property of biological systems to use only the L-enantiomer amino acids.
6. While monolayers formed from pure solutions of bases are crystalline, those formed from *mixtures* of adenine and uracil are *aperiodic* (Sowerby *et al.,* 2000). Such co-adsorption has been confirmed electrochemically (Kirste and Donner, 2001), but interpretation of the co-adsorbates is not trivial. Aperiodicity is an essential prerequisite to convey information (Schrödinger, 1944; Cairns-Smith, 1982). In an abstract way, the aperiodic monolayers resemble nucleic acids except that the pseudocrystalline, sugar–phosphate scaffold is provided by a mineral crystal and the arrangement of bases is in a two-dimensional matrix, rather than a one-dimensional string. The spontaneous self-assembly of such solids from aqueous mixtures of nucleic acid bases evaporated on the surface of crystalline minerals suggests the existence of an organic, nonpolymeric, informational architecture.
7. Significantly, the bases can modulate, in a base-specific way, the adsorption equilibria of selected amino acids (Sowerby *et al.,* 2002). The mapping between bases and amino acids is the signature of a coding mechanism (Yockey, 1992) and suggests that the monolayers could specify the arrangement of chiral amino acids before evaporative peptide bond formation.

In our hypothesis (Sowerby *et al.,* 2000), the spatial arrangement of bases in an aperiodic array could function as a template. The amino acids are expected to be bound to the surface by hydrogen bonds formed between a single H atom of the amino acid ammonium group and the H-acceptors of the underlying bases. Molecular modeling indicates that the amino acid arrays will probably be further stabilized by lateral hydrogen bonds between the carboxylate groups of

the adsorbed amino acids and the ammonium groups of their nearest neighbors (Sowerby and Heckl, 1998). If *coding* can occur through specific interactions of the amino acids with the variable electronic surface formed by the adsorbed bases, the spatial arrangement of bases in the aperiodic structure would function as a template for different patterns of amino acid adsorption.

The roles of the isostructural components of the amino acids in promoting specificity of binding to the base template is a key feature of this proposal. Logically, these components (rather than the R-groups) *must* participate in the primitive "coding" mechanism, since only these interactions can lead to a favorable geometry for peptide bond formation. There is clearly sufficient variation in the electronic configuration surrounding the ammonium and carboxylate isostructural moieties to allow discrimination among different amino acid species (Sowerby *et al.,* 2002). The geometry of the H-acceptors of the base layers would mean that the carboxylate and ammonium residues of the amino acids will be bound in-plane to the base surface in the correct orientation for peptide synthesis (Sowerby and Heckl, 1998), thus increasing the chances of favorable collisions. Dehydration is, of course, required to effect polymerization. Peptide bond formation is not kinetically favored in liquid water and would be facilitated by evaporation on the base monolayer. In this context, the monolayer base "template" would effectively fill a role equivalent to that of the substrate binding site of an enzyme.

Rehydrated peptides would, however, be only weakly adsorbed to the surface because the formation of water during peptide bond formation would remove the surface-bound H-atom of each amino acid that provided the primary mechanism for adsorption. The thermodynamic context of cycles of dehydration and rehydration would thus act as a mechanism for peptide/protein synthesis.

The linearity of peptides is a consequence of the polarized structure of amino acids, which can be linked only in a "head-to-tail" fashion. The amino acids will form a two-dimensional layer whose composition is dictated by the base composition of the two-dimensional base layer to which they are adsorbed, and peptide bond formation would be able to take place in any direction on this planar surface. In the simplest scenario, coding of amino acids and peptide bond formation would occur as a one-step process. Polymerization would thus effect the "translation" of the two-dimensional information source represented by the underlying base matrix to a series of one-dimensional products, and, since many peptide-forming events would occur simultaneously, each two-dimensional template could conceivably encode a number of one-dimensional peptide strings. At the same time, such a geometric template would discriminate between α-amino acids and other interfering compounds such as carboxylic acids, because only α-amino acids would have the correct geometry for peptide bond formation.

The dominant base synthesized in putative prebiotic chemistries is adenine, formed through the simple condensation of hydrogen cyanide (Ferris and Hagan, 1984). The concentration of adenine in the prebiotic ocean was low (estimated to have been of the order of 30 μM), and a means to concentrate it may have been needed (Miller, 1987). Adenine adsorbs strongly to inorganic solids such that the fluid–solid phase equilibrium is far displaced to the solid phase (Cohn *et al.,* 2001; Sowerby *et al.,* 2001a,c). Oxidative deamination of the fluid-phase adenine would also give rise to hypoxanthine (Shapiro, 1995), a purine base with similar adsorption characteristics to adenine (Sowerby *et al.,* 2001a) and with the capacity to form similar self-assembled monolayer structures (Sowerby *et al.,* 2002). Adsorbed adenine monolayer structures exclude water (Sowerby *et al.,* 1998a) and, once adsorbed, would be kinetically protected from hydrolytic decomposition. Mineral surfaces would have been a sink for prebiotically available purines and would have sequestered much of the aqueous-phase yield. A corollary is that adsorption would have pulled the chemical equilibrium in favor of purine synthesis, with a consequent higher yield than might otherwise be expected. The purine base, guanine, is also synthesized in reactions that yield adenine, but to a much lesser extent (Levy *et al.,* 2000). Like its deamination product, xanthine, guanine can also form monolayers (Heckl *et al.,* 1991; Sowerby and Petersen, 1999). Chemistry, it would seem, deems that prebiotically synthesized purine bases would be predominantly displaced to the surface of mineral solids, and one might picture that primordial monolayer composition would be dominated by a mixture of ade-

nine and hypoxanthine, punctuated by occasional guanine and xanthine molecules.

Adenine and hypoxanthine have been, in fact, proposed as the coding elements of a primitive two-base, purine-only coding system (Crick, 1968). Both bases, when adsorbed to a graphite surface, modulate the adsorption equilibria of different amino acids—a process of coding (Sowerby *et al.*, 2002)—and such a modulation phenomenon would be expected to take place on any inorganic solid. It has been proposed that aperiodicity in crystalline inorganic materials had an informational role in the origin of life (Cairns-Smith, 1982). Aperiodicity is not, however, an inherent property of compounds formed by strong long-range electrostatic interactions, because the possible atomic configurations of ionic solids are energetically well separated. Molecular solids, however, are constructed from components that interact predominantly by weak, short-range van der Waals and hydrogen bond interactions, and crystalline polymorphs are possible because different configurations can exist with similar energies (Pertsin and Kitaigorodsky, 1987).

But how could such a system allow for the persistence and evolution of information? A truly aperiodic structure would never repeat; the ability of the information to persist would reside in the substructure of the monolayers and the recurrence of discrete motifs or patterns. Proteins are not continuous but have discrete sizes. A protein-coding portion of monolayer template can be considered as a motif within a larger aperiodic structure, in the same way as coding sequences of nucleic acids are only portions of the genome of an organism. The fact that the tessellation of the mineral surface by the bases will reflect the specific structural rules that dictate interbase hydrogen bond formation would limit the number of "permitted" base patterns that can be formed from a mixture of bases. As a result, the primary structures of the encoded peptides would not be random, but would be expected to comprise a limited number of motifs repeated in different contexts, even though the entire aperiodicity may never be duplicated or replicated. Spontaneous processes such as these circumvent the "chicken and egg" problem of self-replication because no parent template is required. *Reproduction* rather than *replication* of monolayer structure would allow persistence of information. The interparticle interactions are important, because self-assembly is a way of recruiting information that exists within the rules of the interactions. Such self-programmability (Rasmussen *et al.*, 1991) is evident in the self-assembly of crystals from atoms, ions, or molecules and the formation of micelles from lipids, the latter being clearly relevant to the origins of life (Deamer, 1997).

Phenotypic advantage conferred by the catalytic capacity of encoded peptide products that leads to a change in the composition of the monolayer itself would effect an evolution of the information content. In the fluid phase, for example, such a peptide might catalyze the conversion of adenine to hypoxanthine, or *vice versa*, thus changing the surface composition of newly assembled monolayer and, thus, the capacity of the structure to encode new compositions of peptides. The catalyzed synthesis of the other bases would lead to a greater information-carrying capacity and the codes for new peptide catalysts.

In this way, the self-assembly of a G_1-encoded system capable of evolution could lead to a metabolism of encoded catalysts capable of complex organic chemistry, which could ultimately lead to a nucleic acid-based architecture.

This hypothesis is not unlike the crystal gene hypothesis of Cairns-Smith (1982), where aperiodicity in inorganic solids acts as the primordial information source. A distinct difference, however, is the role of heredity in the propagation of the crystal genes. In our model, heredity is not involved; information is held within the bases themselves. Self-reproduction is an inherent property of the monolayers and does not preclude, on the basis of probability, the likelihood of a similar system generating elsewhere in the cosmos where similar conditions prevail. Two attractive features of this hypothesis are its strong experimental basis and the fact that it is amenable to further testing by experiment.

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