# The Origin of Life and the Nature of the Primitive Gene

A. G. CAIRNS-SMITH

*Chemistry Department, The University of Glasgow,*

*Glasgow, W.2, Scotland*

It is proposed that life on earth evolved through natural selection from inorganic crystals.

During the formation of a crystal, certain kinds of lattice imperfections usually replicate as a necessary part of the crystallization process. In so far as those imperfections that replicate are thus self-selecting, any crystallization process is likely to involve a rudimentary biological evolution. Under simple laboratory conditions, such evolution would be limited by the absence of any selection pressure for the elaboration of very complex imperfection patterns. In principle, however, the evolution of such *genographs* could proceed to high levels since, in the form of such patterns, many kinds of crystals could, in principle, hold very large amounts of information.

According to the specific theory that is proposed, the primitive genographs were patterns of substitutions in colloidal clay crystallites. (The theoretical information density in such a crystallite is comparable to that in DNA.) Evolution proceeded through selective elaboration of substitutional genographs that had survival value for the clay crystallites that held them (at first through genetically controlled adsorption of a "spectrum" of organic molecules) within a complex, dynamic, primitive environment. A gradual "take-over" of the control machinery by organic macromolecules—*a genetic metamorphosis*—is then considered to have occurred.

The arguments are based on general considerations leading first to the idea of some kind of genetic metamorphosis having occurred, and then to the conception of the *primitive gene* as a crystal. The specific (silicate) theory follows. Finally, a speculative outline is given for the main stages between the inorganic crystal and the first purely "organic" organisms.

Possible current implications are briefly considered, including a suggestion in connection with carcinogenesis.

## 1. Introduction

### THE INFORMATION PROBLEM

It is the centre of the problem of the origin of life that an organism *seems* to be necessarily far too complex for its chance appearance within a reasonable time and locality under any conceivable circumstances: yet a pre-formed

fully functioning organism of some sort *seems* to be a necessary pre-requisite for organic evolution. In considering one aspect of this problem—the origin of enzymes—Dixon & Webb (1958) have demonstrated the fantastic improbability of even one known protein forming by purely physico-chemical processes. The difficulty is not so much in the appearance of amino acids or of energy, but in the spontaneous appearance of *information*. (Even if the whole earth had been made of nothing but amino-acids which had rearranged themselves randomly and completely ten times a second for the whole period of the earth's history, there would have been little chance of producing even once, for a tenth of a second, one molecule of insulin!)

A piece of dynamite, even if left quite alone, will eventually explode. *Eventually* a chance high energy molecular event will overcome a critical activation energy barrier. The self-propagating process of organic evolution will similarly—in theory—"detonate" spontaneously from an environment containing the necessary components. Here the critical event seems to be the formation of an extremely improbably organized molecular assemblage: here the obstacle seems to be an *information barrier* of prodigious height. (The insulin example gives some idea of the height of the information barrier between the physico-chemical and the modern biological worlds.)

Figure 1 illustrates three general ways in which the information problem may be approached, using:

(1) The idea of spontaneous generation: that the first organisms were simple enough to appear more or less immediately the external conditions were favourable: or,

(2) the idea of "chance and time", as already discussed—that a molecular assemblage, however improbable, will form somewhere, sometime, in an infinite universe. Both of these ideas in their various forms have been discussed, rather critically, by Oparin (1957). Intermediate positions between (1) and (2), such as that expressed by Dauvillier (Oparin, 1957, p. 98) that the spontaneous formation of an organism was of such improbability that it might happen about once in the history of the earth, are unsatisfactory for this reason: there is not very much difference between the degree of organization of a molecular structure that might be expected to appear by chance in about a billion years and one that could be expected to appear almost immediately. Under the idealized conditions for the random synthesis of protein described on p. 59, a given 50-amino acid sequence would be expected to take about a billion years, while a given 40-amino acid sequence should turn up every few minutes!

(3) The idea of abiogenic evolution is the dominant one in most modern theories of the origin of life, and some element of it is an essential constituent in any such theory. Calvin (1953, 1961) and Calvin & Calvin (1964) have
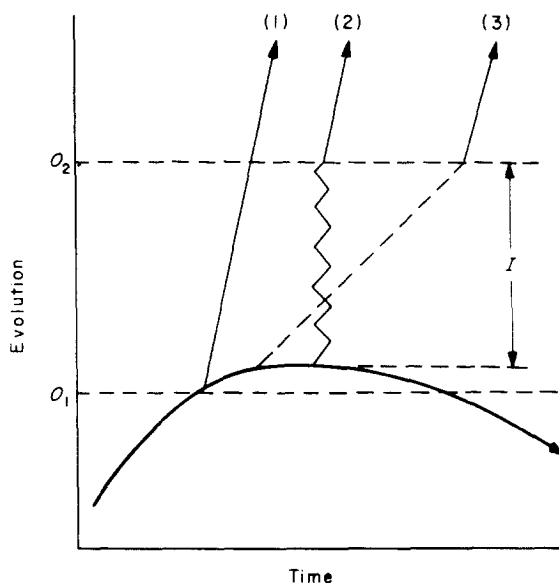
FIG. 1. The broad arrow represents the abiogenic evolution of matter that gave rise to general environmental conditions on the earth. The other arrows represent the evolutions of separate systems within this environment according to different ideas.

$O_1$ is the level of biological information required for the origin of life according to (1) (spontaneous generation). According to other theories, there was an "information barrier" (never less than I) representing the disparity between the amount of relevant information that could be expected to appear in a reasonable time through physico-chemical processes, and the amount required for the simplest possible organism that could evolve to high levels through natural selection. According to (2) ("chance and time") the barrier was jumped by a fluctuation of great improbability: according to (3) (abiogenic evolution) the barrier was climbed through the evolution of systems with only some of the attributes of organisms.

expounded the conception of chemical evolution as part of the general evolution of matter, between the formation of the earth and the appearance of organic evolution, during which relatively complex organic compounds, particularly proteins and nucleic acids, were formed. There is considerable experimental evidence that at least amino acids, random poly amino acids, and nucleotides would have been formed under primitive earth conditions (Miller, 1953; Miller & Urey, 1959; Harada & Fox, 1964; Ponnamperuma, Lemmon, Mariner & Calvin, 1963; Ponnamperuma, Young, Munoz & McCaw, 1964; Fox, 1963).

In his classic work *The Origin of Life on the Earth*, Oparin (1957) discusses very fully various theories on this subject, and follows through in some detail his own evolutionary view. This is summarized in the introduction (p. xii). "Matter never remains at rest, it is constantly moving and

developing and in this development it changes over from one form of motion to another and yet another, each more complicated and harmonious than the last. Life thus appears as a particular very complicated form of the motion of matter, arising as a new property at a definite stage in the general development of matter."

If we think of the universe as being an open system in a steady state—compare the Heraclitean analogy of the universe as a river—then the evolution of matter to which Oparin refers represents a kind of increasingly intricate turbulence in the overall flow (Matter → Radiant Energy?) that drives the universe. The universe then consists of a multiple hierarchy of open systems within open systems. In so far as organisms are necessarily open systems (Oparin, 1957, p. 321 *et seq.*) this kinetic evolution of matter can lead somewhere in the direction of the development of organisms. If we accept the idea of abiogenic evolution completely, then such evolution can indeed go the whole way. On Oparin's theory the final stages were the development of steady-state systems consisting of complex co-acervate droplets maintaining themselves through their chemical activities. Among a population of such systems there would be some which, by virtue of their internal organization, would have greater dynamic stability than others: these would tend to survive more readily, and as time went on more and more dynamic systems would appear. Eventually one such system would acquire the ability to reproduce, and would constitute the first organism: simple selection would have given way to natural selection.

But the change from systems that contained non-replicable information leading to the formation of a stable co-acervate droplet, to systems in which this information was replicable, was still the crucial one. It is not clear that an evolutionary process not involving replication would indeed lead towards the acquisition of this property by the system as a whole. It is not clear that "constant repetition of connected reactions co-ordinated in a single network also led to the emergence of a property characteristic of living things, that of self-reproduction" (Oparin, 1957, p. 360). It may be that such a system could have provided a particularly suitable environment within which some kind of self-replicating control device—a virus-like subsystem perhaps—could have arisen, but the appearance of such genetic machinery would still have represented the crucial stage in the onset of evolution through natural selection: there seems to be every theoretical advantage in this machinery having been simple, and having appeared early.

Oparin (1957, pp. 95–99) has criticized theories, such as that of Muller (1947,1955), that describe the origin of life in terms of the origin of the gene, on the grounds that such ideas are scientifically barren—that there can be no hope of confirming or refuting an occurrence of such great improba-

bility as the spontaneous formation of a gene. It is the purpose of this paper, however, to pursue such a "genic" point of view, to accept as a consequence of this view that the spontaneously forming *primitive gene* must have been a relatively straightforward physico-chemical system, and to attempt to deduce the general structure of such a gene from the known abstract requirements of biological control mechanisms in general and of genetic structures in particular. We will proceed through a series of increasingly specific questions.

## 2. General Theory

### Q.1. WERE THE PRIMITIVE MOLECULAR CONTROL MECHANISMS SIMILAR TO THOSE IN MODERN ORGANISMS?

A detonator, an egg, and a starter motor have this in common: they are each a special device used to overcome a starting problem. Once the process that they have helped to initiate is under way they can be discarded. The problems of starting are often quite different, and so require different structures, from the problems of further development. If ever a process
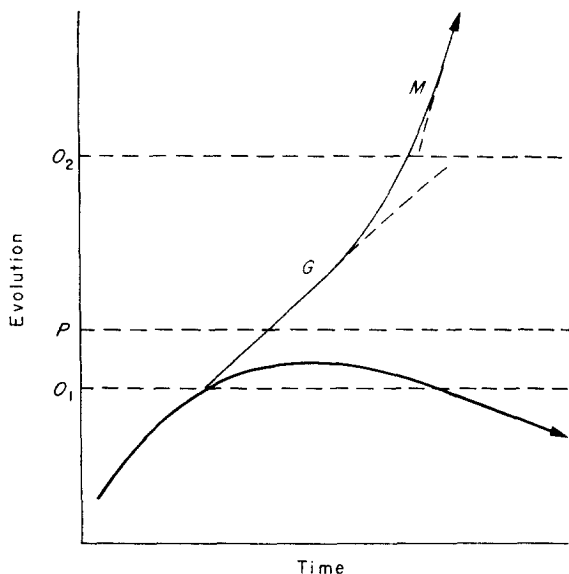


FIG. 2. (Compare Fig. 1.) According to the fourth idea, of genetic metamorphosis, a primitive life form originated spontaneously at $O_1$ and evolved through natural selection. (The genographic theory regards the hereditary machinery in such organisms as having been (inorganic) crystals.) A "take-over" of the genetic machinery (by organic polymers) may have started quite early (say at $G$), and was complete at some point at or beyond $O_2$ (say at $M$). $P$ is the kind of information level that might represent a single insulin molecule, and is the kind of level that may be achieved at present by laboratory synthesis.

seemed to need a special starting device, it was organic evolution. For this process the "starting problem" was largely to be simple enough; the "problem" of evolving to high levels involved the opposite requirement of great complexity. On these grounds alone we might expect the very first organisms to have been totally different from modern organisms. We might expect to find, in particular, that their molecular control machinery worked on a very different principle from that of the modern universal DNA/RNA/ protein system: that they used not just a simplified form of the modern bio-cybernetic machinery, but a quite different "self-starting" system. (Compare a four-stroke engine which does not become self-starting by simplifying the design—it requires another device to start it that works on a different principle.)

Figure 2 illustrates, then, a fourth general way in which the information barrier, referred to in the last section, could have been overcome—through a process of *genetic metamorphosis*. According to this idea, there were, in effect, two origins of life:

(1) the origin of primitive life, simple enough to have appeared spontaneously, and to have started the consistent creation of biological information through natural selection;

and:

(2) the origin of "modern" organisms, flexible enough to be able to evolve to very high levels. (There may, of course, have been several "gear changes", but this is the simplest form of the idea.)

## Q.2. WERE THE PRIMITIVE CONTROL MECHANISMS BASED ON LINEAR ORGANIC POLYMERS?

The discontinuity introduced by a process of genetic metamorphosis could effectively conceal the primary origin of life in any attempt to extrapolate from known organisms to primitive forms. (An examination, however detailed, of a working four-stroke engine might give no clue as to the exact structure of the mechanism by which it was started—one could only state general requirements.) We know only that we are looking for a self-starting life form, for a form that could appear as a consequence of the inherent self-organizing properties of matter at low information levels—the kind of matter that is familiar to the physical chemist.

DNA does, of course, replicate under suitable conditions, and it is quite possible that replicability may prove to be a property of some much simpler kinds of polymer systems: it is indeed possible to envisage a polymerization process, analogous to a process of crystallization, in which sequential information is replicated (Pattee, 1961). But such systems are not common:

replicability is by no means a characteristic property of linear organic polymers: replicability is, on the other hand, a characteristic property of simple crystals. A growing crystal represents, indeed, a naturally occurring "Penrose machine" (Penrose, 1959)—a point that is emphasized when we consider actual crystal growth mechanisms (see section Q.4, e.g. Fig. 6). Again, information to be effective must presumably operate through reasonably specific molecular interactions: such specificity is more often found on the surfaces of simple crystals than on *simple* organic polymers.

Q.3. WHAT WERE ESSENTIAL GENERAL PROPERTIES FOR PRIMITIVE GENES?

They had to be self-forming under relatively simple physico-chemical conditions. They had to hold information. They had to replicate information. They had to be able to communicate the information that they held to their immediate environment. They had to be able to evolve. These requirements can be expressed in terms of seven general properties for a primitive genetic material.

(1) *Low intrinsic information content.* For primitive genes to have arisen under simple conditions, the information inherent in their structure, and in any necessary associated machinery, must have been small.

(2) *A potential ability to hold relatively large amounts of information.* The primitive genetic information had to be able to evolve to around $O_2$ in Fig. 2; hence also:

(3) *Stability.* The information had to be maintained (for perhaps millions of years). (See also (5).)

(4) *Ability to replicate information.*

(5) *Accuracy in replicative function.* This is a kind of dynamic stability; it is perhaps the most critical feature of all. By the time the information had evolved to a level equivalent of 1000 units of a binary code (about the amount needed to code a small protein), then the equivalent of 99% accuracy in the copying of each unit—and such accuracy is regarded as "good" for a synthetic organic reaction—would result in an average of ten mutations per replication. This would probably be too much for the selection processes. Reasonable accuracy *in its replicative function* seems to be an intrinsic genetic requirement: it is difficult to see how it can be produced by Darwinian evolution, because it is difficult to see how Darwinian evolution could go very far without it.

(6) *Mutability.* In spite of (5) occasional arbitrary changes in the information held must occur and be inheritable.

(7) *The information that it holds must have survival value for the gene*; i.e. it must be biological information. This involves a great variety of possible *communicative* functions, depending ultimately on information-dependent interactions of the gene with its own immediate environment. This communicative requirement will be dealt with in Q.5 and subsequently.

### Q.4. WHAT WAS THE GENERAL PHYSICAL STRUCTURE OF THE PRIMITIVE GENE?

*Crystallization* is, in general, a spontaneous and accurate replicating process and some crystals, such as diamonds or silicates, can be very stable. The crystal has long been recognized as an organism analogue. If one thinks of perfect crystals the analogy is limited by the trivial amount of information being processed (Moore, 1964). But real crystals are always imperfect, and in such imperfections large amounts of information could, in principle, be held. Some simple crystal, then, might have at least the primitive genetic requirements (1) to (6) above, provided that it could contain stable patterns of imperfections that replicated, with occasional "mistakes", during crystal growth. We will pursue this hypothesis further, in the light of ideas on imperfection structure and growth mechanisms in crystals, which will now be outlined briefly.

### Notes on imperfections and growth mechanisms in crystals

(See, e.g., *Discuss. Faraday Soc.*, 1949; "Growth and Perfection of Crystals", 1958; Gilman, 1963.)

#### (i) Physical imperfections

Insofar as any real crystal is not infinite it is "imperfect". Thus the size of a crystal, the relative growth along different axes (crystal habit), or a distribution of size or habit in a group of crystals, all represent ways in which a crystalline sample could contain information. Information could be "written" as a pattern of grain or domain boundaries in a compact crystalline material. (The information in a magnetic tape is something of this kind.)

Point defects are localized lattice imperfections—commonly vacancies. They are usually thermodynamic features, i.e. essential constituents of the stable crystal at normal temperatures; for this reason they are not likely to be of interest to us.

Simple dislocations are metastable features commonly associated with the processes of crystal growth; they fall between the extremes of the grain boundary, which is a mis-match of two separate lattices, and the point defect, which is an isolated imperfection in an otherwise perfect lattice. An edge dislocation, for example, results from a slip at right angles to a line

defect within a crystal, and a screw dislocation to a slipping of lattice units parallel to a line defect. Figure 3 illustrates these kinds of physical imperfections.
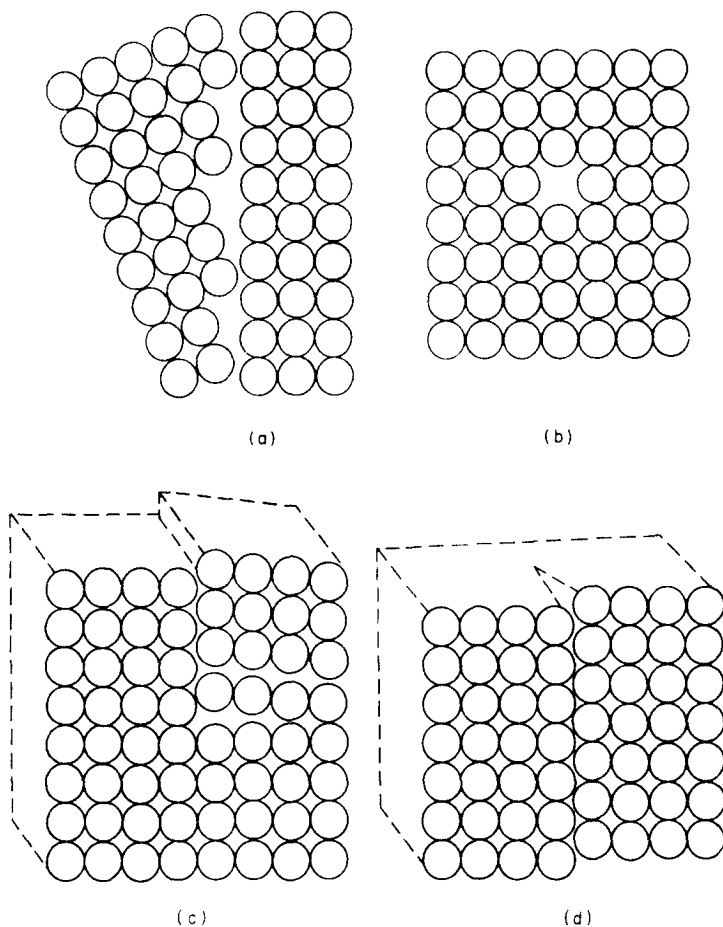


(a)

(b)

(c)

(d)

FIG. 3. Physical imperfections (in crystals) which are discussed as potential information elements.
  (a) Grain boundary; (b) point defect; (c) edge dislocation; (d) screw dislocation.

Complex systems containing many such imperfections may occur.

### (ii) Chemical imperfections

These may be *interstitial*, where "foreign" atoms or groups are found in spaces between normal lattice constituents—e.g. carbon in steel; or

*substitutional*, where foreign atoms or groups replace some of the normal lattice constituents—e.g. arsenic in germanium (see Fig. 4).
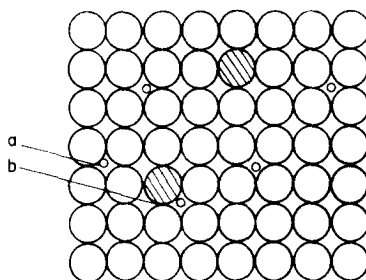


FIG. 4. Chemical imperfections (in crystals) that are potential information elements. a. Interstitial; b. substitutional.

### (iii) Complex imperfections

By introducing strains into the lattice, chemical imperfections may give rise to physical ones, and, by creating special adsorption sites, physical imperfections may attract specific "foreign" components. (In section 3 we will consider a particular kind of complex imperfection as having been the crucial one; here we will continue with simpler and better understood systems.)

*Crystal growth mechanisms.* The growth of a crystal face through random cohesion of individual molecules, or through simple layer-by-layer addition of molecules, is easy to imagine but probably wrong—at least for most crystallization processes at relatively low supersaturation levels. The cohesion of an isolated molecule on a perfect surface (Fig. 5, a) is very much lower than the cohesion in a "step" (Fig. 5, b) or "corner" (Fig. 5, c). Growth
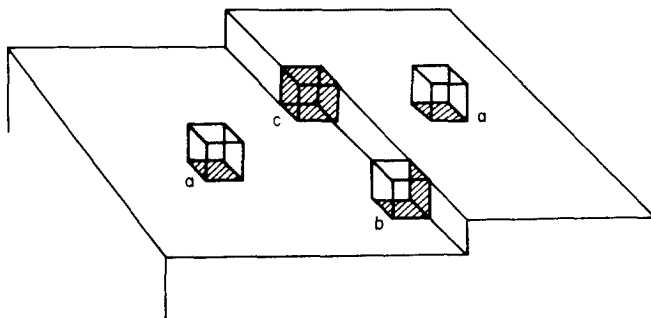


FIG. 5. Growth of a crystal face tends to occur by the migration of "steps". (The cohesion of a molecule c $\gg$ b $\gg$ a.)

tends to occur through the migration of steps (Figs 5 and 6), and to depend on the pre-existence of such steps. These may arise through imperfections of the kinds discussed. Growth at an edge dislocation illustrates the basic idea (Fig. 6). Here growth occurs *because* the dislocation propagates through
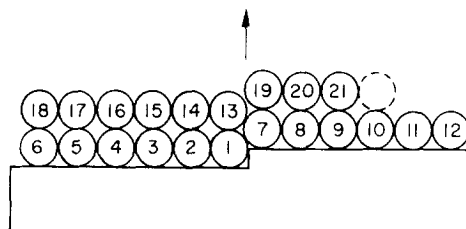


FIG. 6. The replication of a dislocation may provide a mechanism for rapid crystal growth.

the crystal. If the addition of new molecules could create a perfect surface at any time, i.e. if the dislocation could be eliminated, then the growth of the crystal would virtually stop. Growth on the spiral ramp generated by a screw dislocation similarly depends on the concurrent propagation of the dislocation. In crystalline samples certain kinds of imperfections, then, will owe their prevalence to the fact that they were self-propagating—their existence was kinetically favoured through an orderly process of crystal growth. Further, dislocations, patterns of dislocations, or grain boundaries present in a seed crystal may be inherited by the growing crystal, e.g. in the growth of silicon (Dash, 1958) or alkali halide crystals (Dobrzhanskii & Shaskol'skaya, 1964).

As pointed out by Frank (1949) "a rather odd natural selection" operates in the kinds of crystal growth mechanisms that we have considered. Only some imperfections produce growth through their own propagation, and these will tend to be by far the most prevalent in a crystal, with the most "efficient" the most prevalent. They are selected for their replicative efficiency. There is no reason to suppose that such selection will generally favour particularly complex patterns of imperfections, so evolution along these lines would normally be limited to the production of relatively simple "information" in the form of more or less separate screw dislocations, etc. But complex patterns might be favoured if these could have an effect on the immediate environment of the crystal holding them, so as to increase the chances of that crystal surviving in competition with others; if, in short, the primitive genetic condition Q.3. (7) could be satisfied. Evolution could then proceed further.

Q.5. WHAT WAS THE NATURE OF THE IMMEDIATE CONTROL FUNCTION
OF THE PRIMITIVE GENE?

Taking a "genic" view of modern organisms, we can think of them as consisting of the molecular patterns—probably DNA base sequence—that constitute their genetic information. The body of an organism is then a kind of (surprisingly!) elaborate device for improving the chances of survival of a particular library of sequences, by converting a relatively hostile external environment into a special immediate environment in which the DNA can survive and replicate. DNA is not really a self-replicating molecule, it requires an environment of high information content in order to replicate, and its control function—in creating and maintaining a "body" for itself—requires the pre-existence of a "body". (An individual develops, not from a set of DNA sequences, but from a zygote—which already has a "body".) But if the view which is being pursued here is right, the primitive gene could survive and replicate without a "body" at all. A crucial question is whether such a gene could improve its chances of survival by replicating a pattern that caused it to acquire a "body", and whether such a pattern could be simple enough to arise by chance.

Considering again modern organisms, the interaction of DNA with its own immediate environment is a *pattern-dependent interaction*. The genetic information *is* a pattern, and the whole decoding mechanism is a device that gives this pattern significance—through the synthesis of specific proteins to the construction of enzymic pathways, cell structures, and so on. The genetic information in the first genes, however, had to be far more immediately significant: it must have worked through some very straightforward physico-chemical pattern-dependent interaction. Such an interaction is the more or less specific "fitting" of two molecular surfaces. In modern organisms, specific "plug and socket" interactions represent a basic molecular control technique: here one can often distinguish between the "plug" and the "socket", much of the significance of protein being its ability to form specific "sockets"—a property rare in molecules, and virtually unknown in molecules formed under physico-chemical conditions. The surface of a crystal, however, is a "socket-structure", and physical adsorption of molecules on a solid surface can be a "plug and socket" kind of interaction. The adsorptive, and indeed catalytic, properties of a crystalline surface may be strongly affected, if not dominated, by the presence of imperfections emerging at the surface (Vol'kenshtein, 1963). It is possible to imagine a crystal in contact with a mixture of dissolved substances adsorbing a particular population of molecules that depends on the imperfection patterns that the crystal contains. In crystallites of colloidal dimensions, the nature of adsorbed molecules and

ions play a dominant part in determining the physico-chemical properties of the system—whether the particles remain dispersed, flocculate or form a gel, for example, can depend critically on the molecular or ionic species that the particles tend to adsorb: clays may even change from being dispersible in water to being dispersible in oil through the adsorption of polar organic molecules (van Olphen, 1963, p. 179).

We are now in a position to suggest a general answer to Q.5. "The primitive genes, or rather the very early primitive genes, controlled their immediate environment through adsorption of molecules present in the environment." We can now envisage a more highly evolved organism than the simple imperfection-containing crystal. Such an "adsorption organism" is a colloidal crystallite (the gene), with a more or less genetically controlled population of adsorbed molecules (constituting the rather vague "body" of the organism). In so far as such a population of adsorbed molecules can affect the physico-chemical properties of the system, it is possible that under particular natural circumstances—and such will be considered in section 4—such a "body" could be biologically efficient, i.e. tend to preserve the crystallite holding it.

There would be two stages in the replication of such a crystal-gene organism: crystal growth, involving the internal replication of imperfection patterns, and crystal cleavage, by which the internal information would be exposed on the surfaces of the crystals. This second stage presents no great difficulty since crystals normally break up during growth, particularly if they contain many imperfections—the large crystal is a relative curiosity.

### The general (genographic) theory of the origin of life

This is a form of the even more general idea of genetic metamorphosis. The theory proposes, in effect, the existence of intermediate forms between simple crystals, on the one hand, and modern organisms on the other.

The key idea is that the primitive genetic information was held not as a pattern in a *molecule*, but as a kind of "picture" of imperfections in a *crystal*. Such an imperfection pattern we will call a *genograph*.

### Comment

Even without being more specific, we can see that this rather abstract entity—the genograph—could in principle be self-forming and self-replicating through the processes of crystal growth, and self-selecting through (at first) simple adsorptive environmental interactions. (The ability to bring about catalytic conversions in the adsorbed molecules might well be advantageous, but evolution could have been under way before any such subtlety emerged.) What is required, after all, is not necessarily a self-reproducing

*molecule*, but a self-reproducing control device of some sort: in the physico-chemical world from which life emerged, replication and molecular control are more typically crystalline than molecular functions.

## 3. Specific Theory

Q.6. OUT OF WHAT SUBSTANCE WAS THE PRIMITIVE GENE MADE?

The picture of the primitive gene that has so far been developed would probably fit a large number of different substances. The need for specific adsorptive interactions with the environment—to give significance to rela-tively complex imperfection patterns—suggested crystals of colloidal dimen-sions, and this in itself suggests an inorganic material that is rather insoluble in water. As the material must have formed under geological conditions, what we are probably looking for is a mineral. "A colloidal mineral" suggests a *clay*. As we shall see there are several other arguments that lead in this direction. Bernal (1951) suggested that clay particles could have provided sites for the concentration, ordering and catalytic reaction of simple organic molecules that were probably present on the azoic earth. This idea was used by Bernal to account for the formation of the more complex organic molecules that have been generally assumed to be necessary precursors of life. On the view being pursued here the significance of the clay particles could have been somewhat different and even deeper: perhaps they were not simply the sites where organic molecules could congregate and react to give products that in their turn gave rise to organisms: perhaps the clay particles *were* the first organisms, and the early evolution of life consisted of the elaboration of information *in* clay particles—as the much later evolution was to consist of the elaboration of information in DNA.

There is a particularly elegant way in which clays, and silicates in general, could hold information. The silicate minerals can be thought of as complex condensation products of silicic acid formed into fibrous (e.g. asbestos), layered (e.g. mica, clays), or three dimensional framework structures (e.g. feldspars). (Between them these kinds of crystalline giant molecules consti-tute most of the solid material in the earth's crust.) The variety of such minerals derives largely from the possibilities of substitution of silicon atoms by other atoms—e.g. aluminium. In many silicates, and particularly among clays, substitutions of these and other kinds—see later—can be arbitrary within limits, a range of compositions, for example, being possible. Usually, as with aluminium for silicon, the substitutions give rise to negative charges in the structure which are neutralized by more or less independent cations —such as $K^+$ or $Ca^{2+}$. In such substitutions, within an otherwise highly organized crystalline structure, there is a potential very stable and virtually

unlimited information storage capacity. (Assuming one arbitrary substitutional feature per unit cell of a clay, the maximum information density (by weight) would be about half that of DNA.)

## Notes on clay structures

(See also, for example: Pauling, 1960, pp. 553–562; Huckel, 1951; Iler, 1955; van Olphen, 1963.)

Si—O bonds have at least some covalent character, but it is often useful to think of silicate minerals as purely ionic crystals in which the anions are mainly large $O^{2-}$ ions that take up most of the space and are more or less closely packed together with (usually small) high valence cations—$Si^{4+}$, $Al^{3+}$, etc.—fitting into the spaces left between the oxygen spheres. In clays
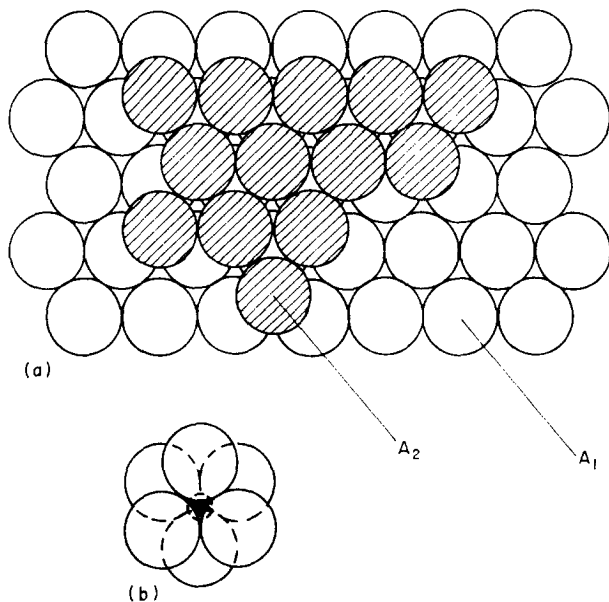


FIG. 7. Simple close packing of two layers of spheres (a), gives rise to "octahedral sites" which are surrounded by six spheres (b).

this gives macro-anionic sheets which then associate with (often) larger and lower valence cations.

Consider a single close packed layer of spheres. Call this "layer $A_1$". One may now add another identical layer that is displaced laterally, but with the same orientation as $A_1$. Call this "layer $A_2$" (Fig. 7(a)). Another way of adding a second layer would be with the rows of spheres rotated by 30°, but to do this the spheres would have to be slightly smaller and, if this second

layer is to be quite flat, every second sphere in every second row would have to be omitted. This would give a hexagonal net arrangement (Fig. 8(a)).
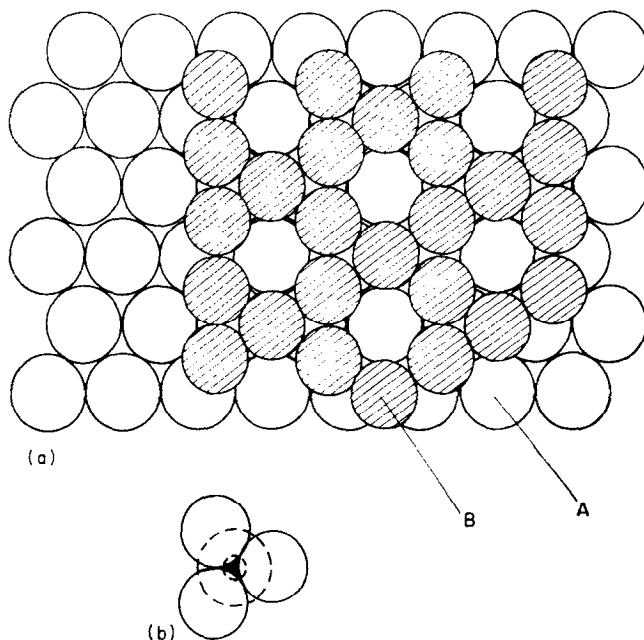
FIG. 8. A flat hexagonal net of smaller spheres may be laid on a close packed layer of spheres as in (a) to give "tetrahedral sites" (b).

Call such a layer $B_1$ or $B_2$ according to whether it is lying on $A_1$ or $A_2$, respectively.

We are now in a position to give a general description of the two main types of layer-lattice silicates. *Two layer silicates* (e.g. kaolin) have two layers of cations sandwiched between three layers of $O^{2-}$ ions, these latter being stacked in the sequence $A_1$—$A_2$—$B_2$. In *three-layer silicates* such as mica or montmorillonite, three layers of cations are held between four oxygen layers which are stacked in the sequence $B_1$—$A_1$—$A_2$—$B_2$. The cations between the two A layers are in "octahedral" positions—i.e. they have six oxygen atoms surrounding them (Fig. 7(b)), those in the positions between A and B layers are in "tetrahedral" sites—i.e. they are surrounded by four oxygens (see Fig. 8(b)). The structure of a three-layer silicate, then, could be represented more fully thus:

$$B_1—t_1—A_1—0—A_2—t_2—B_2.$$
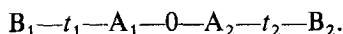
In three-layer silicates, one in three of A layer oxygens are hydroxyl groups.

In the uncharged three-layer silicates talc and pyrophyllite all the tetrahedral sites are occupied by silicon while the octahedral ones are fully occupied by magnesium (talc), or two-thirds of them are occupied by aluminium (pyrophyllite). Substitutions by lower valence metals may be in the tetrahedral, octahedral, or both classes of positions to give negatively charged sheet structures which together with other "independent" cations may give a typical three-layer clay particle. This is illustrated in the sketch (Fig. 9).
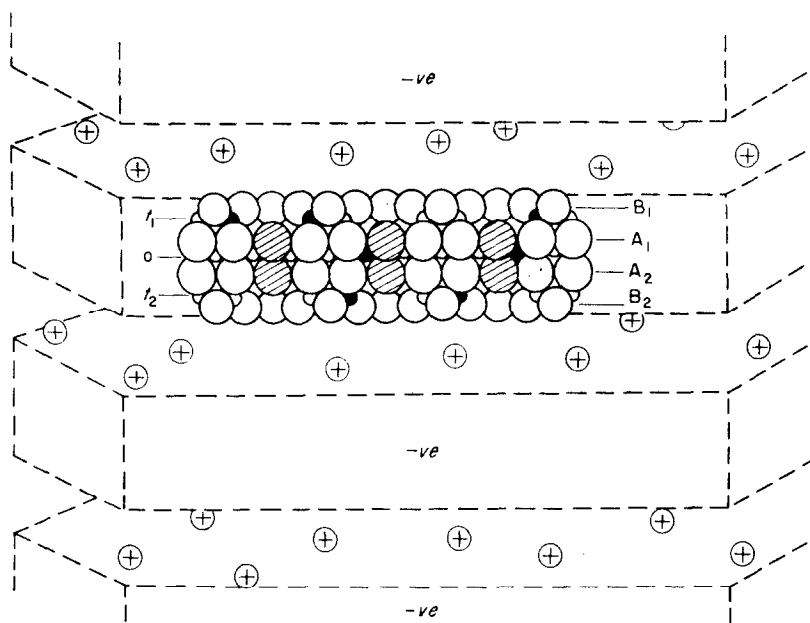


FIG. 9. Sketch of part of a three-layer silicate crystallite showing internal cation substitutions (● for ○) that give rise to negative charges in the sheets, and which may have been the elements of the primitive genetic information store.
○, $O''$ ions; ⊙ and ●, intra-layer cations, ⊕, inter-layer cations; ●, $OH^-$ ions.

### Possible pattern-replication mechanisms in clays

Clays may be formed by alteration of other silicates (diagenesis) or directly from solution (genesis) (van Olphen, 1963, p. 78). The processes may take place at high temperatures and pressures—hydrothermal synthesis —or from very dilute solutions very slowly at normal temperatures and pressures (see "Genèse et Synthèse des Argiles", 1962).

Although clays can be synthesized in the laboratory, it is not surprising that the processes of clay synthesis are not yet well understood in view of

the difficulties associated with even simple crystallization mechanisms. Under hydrothermal conditions it may well be that the structures formed represent something approaching thermodynamically stable types (Roy, 1954), but under normal conditions the very slowness of the process suggests that kinetic factors are important—that the structures formed will depend strongly on the existence of mechanisms for their formation.  According to Caillere & Henin (1962) the pre-existence of some degree of octahedral organization is required for clay synthesis under normal conditions. Thermodynamic reasons for believing that some kind of pre-organization is essential have been discussed (Fripiat & Gastuche, 1962). Whatever kind of "seed" is required it seems likely that clay synthesis from solutions proceeds through some kind of nucleation and growth process. If this is so, the pre-existence of imperfections in the seed is likely to be relevant to the
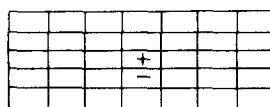


(a)
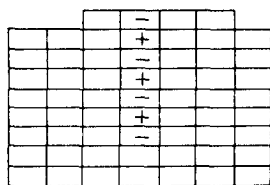


(b)

FIG. 10. Hypothetical process that could duplicate (a) or multiply (b) substitutional information elements during the growth of a crystal.

growth mechanisms as it is to other crystallization processes. A substitution —Al for Si for example—can produce two powerful effects; by introducing a charge and by distorting the lattice (e.g. Si—O bond length is about 1·61 Å while the Al–O length is about 1·78 Å, Pauling, 1960, pp. 321, 359). The subsequent growth of a crystal containing such a substitution must involve the local neutralization of the charge, if Pauling's electrostatic rule is to be obeyed (Pauling, 1960, p. 548), and it will tend to relieve lattice strain if possible. Compensation may be possible by a second substitution (Fig. 10(a)) or alternatively a cyclical sequence of processes could be set up that re-introduces the original substitution, resulting in the propagation of a line of substitutions through the crystal. Figure 10(b) illustrates such

a hypothetical process (compare the propagation of an edge dislocation during crystal growth, Fig. 6) other similar mechanisms, for example, propagating a "large–small–large . . ." sequence, can be envisaged. If we now imagine a two-dimensional *pattern* of initial substitutions, such a pattern could either simply duplicate a "negative" version of itself, or "print" alternative "positives" and "negatives" (Fig. 10(a), (b)).

A recent investigation of the detailed structure of muscovite (Gatineau, 1964) suggests that some such replication process occurs during the formation of this layer-lattice silicate. (Muscovite (mica) represents a relatively easily studied prototype for clays, having a typical three-layer structure, but having crystals that are large and well formed. A typical composition approximates to $Si_3^{4+}$, $Al^{3+}$ (tetrahedral); $Al_2^{3+}$ (octahedral); $O_{10}^{2-}$, $(OH)_2^-$ (anions); $K^+$ (interlayer cations compensating the tetrahedral Al substitutions).)

According to Gatineau (1964), from a study of diffuse X-ray patterns produced by a muscovite of this type, the tetrahedral Al substitutions in this mineral are arranged in zig-zag parallel rows, and the crystal is divided into principal domains in each of which the rows are in one of three possible directions. (There are also indications of minor domains with different orientations.) In addition, a complete Al row must be separated by at least one Si row: the domains are thus divided into *bands* (involving typically 2, 4, 6, or 8 rows of tetrahedral layer hexagons) that are effectively half-substituted, separated by unsubstituted bands. The need for local charge balance seems to give rise to a most interesting feature: *between two adjacent layers, an unsubstituted tetrahedral band comes immediately above a half-substituted one and vice-versa.* The arrangement is illustrated in Fig. 11. The very constant proportion of tetrahedral substitutions in micas—and that it should be one in *four* in a *hexagonal* net was rather a puzzle (Rado-slovich, 1960)—is thus explained as arising from a vertical pairing of unsub-stituted with half-substituted bands. The domain structure on one face of a mica sheet is thus copied—with the band structure reversed—on the adjacent sheet. This could provide the crucial element of a replicating process. Since it is the crystal as such that is divided into domains, the pattern in one set of tetrahedral position in a given layer is presumably also copied in the other set of tetrahedral position in the same layer.

While the above structure for muscovite suggests that a duplication or multiplication process occurred during the formation of the crystal, its importance here is more in illustrating the general point that layer-lattice silicates can have a structure that is to some extent arbitrary *within* the layers—here the pattern of banded domains—and highly ordered *between* the layers. Inter-layer ordering is also indicated in the chromium chlorite

studied by Brown & Bailey (1963). Considering this question generally, any crystal, if it is an information replicating machine, should have some kind of "semi-ordered" structure: there should be some structural *disorder* in the planes or lines that contain the information (compare the "disorder" in the
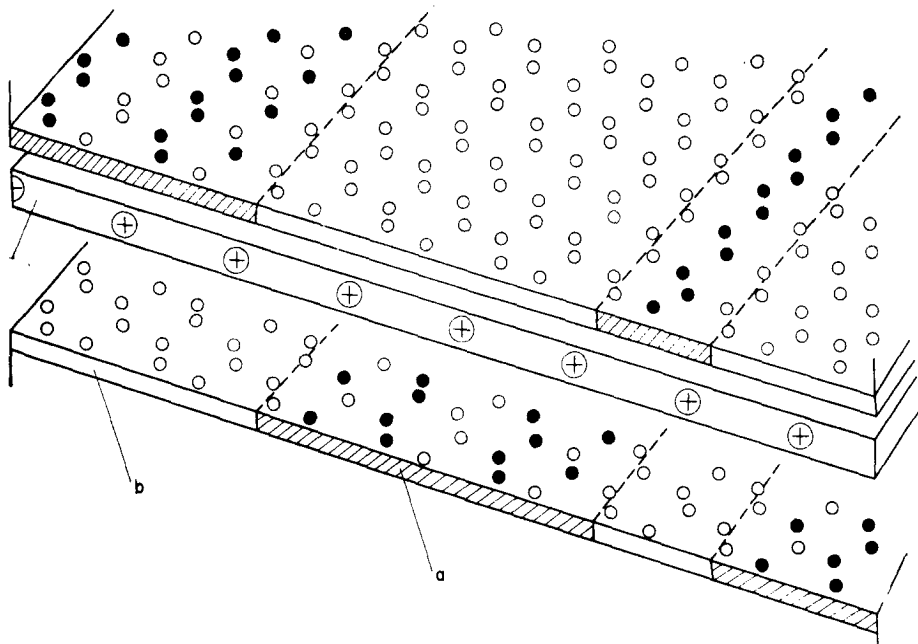


FIG. 11. Tetrahedral cations in two adjacent sheets of a single domain in muscovite, and an arrangement of these cations into parallel half-substituted (a) and unsubstituted (b) bands. Between the sheets there is an ordered array of $K^+$ ions (c) neutralizing the Al substitutions (●) *either* above or below. The band structure (and domain structure) in one sheet is thus copied in reverse in the adjacent sheet (Gatineau, 1964). If such a copying occurred during the growth of this, or some similar silicate crystal, it could have provided an information replicating mechanism for primitive organisms.

○, Si; ●, Al; ⊕, $K^+$.

sequence of DNA bases), and structural *order* in the direction or directions of crystal growth (compare the accuracy with which DNA base sequences are copied). This combination of *static disorder* with *dynamic order* is a characteristic of simple replicating processes of more general kinds—e.g. printing.

It would be premature to identify mica—or indeed any three layer silicate— as the primitive genetic material. It remains to show, for example, that ordering of the kind that occurs in mica is, as it seems to be, kinetic (e.g. that it results from some nucleation and growth process), and that some such

"semi-ordered" layer-lattice silicate can form under "biological" conditions (e.g. by genesis from solutions at reasonable temperatures†).

Ordering between octahedral substitutions might provide a mechanism for information replication in montmorillonites—where substitutions tend to be predominantly octahedral. Figure 12 illustrates the rather simple kind of arrangement envisaged.
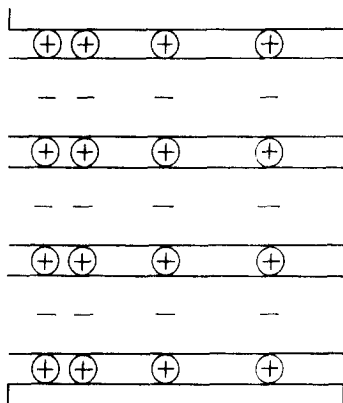


FIG. 12. Replication of information held as a pattern of octahedral substitutions in a layer-lattice silicate might be possible through simple interlayer ordering.

In layer-lattice silicates, the final stage of a replication process—the partial or complete separation of the information-containing layers—can be particularly easily envisaged. Such separation can occur through changes in external conditions—for example even mica "swells" in the presence of n-alkyamonium ions (Weiss, 1963). (Indeed Weiss, 1963, p. 222, has suggested that such intracrystalline swelling of clays may be of interest to biochemists in as much as it is a simple model for self-reproduction.) In nature, cyclical

† It is conceivable that in the formation of clays under normal conditions from dilute solutions, the layers form independently—the three-dimensional crystal appearing subsequently by combination of these layers with cations. A rather different crystal replication process (reminiscent of DNA) involving copying of tetrahedral substitution patterns, could still, however, operate—at least in the formation of a three-layer clay—through the splitting of the layers along the central octahedral plane with concomitant or subsequent synthesis of new half-layers on the half-layers so formed. (The octahedral bonding is more ionic than the tetrahedral.) Diagenetic processes where octahedral cations are exchanged for other cations suggest that some octahedral separation can occur in clays, while the idea of Caillere & Henin (1962), that octahedral pre-organization is required for clay synthesis, would also fit in with such a duplicating mechanism. This is highly speculative, but it perhaps illustrates at least that genetic functions are more easily imagined in natural *layer* silicate structures than in those kinds of *linear* organic structures that form outside organisms.

and sporadic variations in ambient conditions are usual (e.g. diurnal temperature changes, changes in weather conditions, etc.) and these could in principle drive a replication process of the kind envisaged. (As already remarked (p. 60) crystals tend to break up anyway during their formation, so the problem is not a very critical one.)

### Notes on general interactions of clays

Only the barest outline of the prolific variety of clay interactions can be given.†

Clay systems may be in a suspended, flocculated, or gelatinous state, depending critically on interactions between the clay crystallites themselves and on interactions between the crystallites and other components. The individual layers out of which the crystallites are formed may be in relatively close contact, or the systems may be "swollen" through the uptake of water or organic molecules between the layers. The layers may also be more or less completely separated from each other. At layer and crystallite edges there are probably positive charges due to exposed octahedral and tetrahedral cations (van Olphen, 1963, p. 90), so that "card house" structures may be formed by association between the edges and (negatively charged) faces of particles.

Clays interact more or less specifically with inorganic cations (on and between layers, e.g. metals, $NH_4^+$); inorganic anions (at edges—polyphosphate is particularly active); organic cations (aliphatic, aromatic, heterocyclic); organic anions (particularly tannates and other oligo-anions); polar organic molecules (e.g. glycols, ketones, amino acids); polar and ionic organic polymers (e.g. proteins, polyacrylates). Such interactions may be very sensitive: the addition of very small amounts of a polyelectrolyte (tenths of a per cent) may protect a suspension against flocculation, while an even smaller amount (thousandths of a per cent) may have the opposite effect. These phenomena are probably due to physical envelopment and cross-linking respectively (van Olphen, 1963, p. 170).

The interactions of layer-lattice silicates with organic molecules can be highly ordered, a kind of two-dimensional crystal of organic molecules being formed between the silicate layers. Many heterocyclic and aromatic bases were found to stack with their planes either parallel to, or at right angles to, the layers in montmorillonite, the orientation depending on the concentra-

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† See, however, van Olphen (1963), Iler (1955) and the series of volumes "Clays and Clay Minerals, etc." published since 1954; particularly the proceedings of the *ninth* conference (1962) on Clays and Clay Minerals which features symposia on clay–water relationships with respect to engineering properties of soils, and on clay–organic complexes. In the proceedings of the *tenth* conference (1963) another symposium on clay–organic complexes is featured.

tion of the organic component (Greene-Kelly, 1955). From infra-red studies of complexes between ketones and calcium montmorillonite, it has been suggested that the clay surfaces act as nuclei for the two-dimensional solidification of the organic components (Tensmeyer, Hoffman & Brindley, 1960). Weiss (1963) has prepared, and characterized by X-ray and chemical methods, more than 8000 different derivatives of "mica-type" layer silicates (muscovite, biotite, illite, vermiculite, beidellite, montmorillonite), with $N$-alkylammonium ions and other organic compounds. These organic compounds can form what we might call "two-dimensional mixed crystals" between the silicate layers, e.g., where one of the components is $N$-alkylammonium and the other the corresponding alcohol.

Weiss (1963) has also considered catalytic reactions involving alkylammonium ions within layer-lattice silicate complexes, in particular the hydrolytic cleavage of proteins, which may occur at regions of high charge density, and also the oxidation of aniline.

The state of oxidation of Fe in chlorite may be changed without destroying the crystal structure, and it is possible that much of the $Fe^{3+}$ reported in analyses were originally $Fe^{2+}$ which were oxidized with loss of H from hydroxyl groups (Albee, 1962). Here we see the possibility of clay structures acting as hydrogen carriers.

Finally it is perhaps worth noting that clays are important industrial catalysts (100,000 tons being used by the petroleum industry in 1956 (Ryland, Tamele & Wilson, 1960, p. 1), as catalysts for cracking and reforming reactions).

### Possibility of pattern-dependent interactions in clays

We are supposing that the primitive genetic clay could perform both the general functions of DNA and of protein; that the genograph was not only self-replicating during clay synthesis, but that it could also directly control the molecules around it. To know what kinds of genograph-dependent interactions could have survival value for the clay particles, would involve at least a detailed knowledge of environmental conditions. This will be considered in the next section along with other environmental considerations—sources of energy and materials.

One should not necessarily expect to find evidence in the literature for specific genographic effects in clays (as one should not expect specificity in the interactions of random poly amino acids): modern clays are probably "non-biological", and so specific effects will not have been selected for. But there are some indications that, at least in principle, specific surface effects could arise as a result of particular substitution patterns in clays.

To use an analogy with proteins, we can see that enzyme specificity is in

principle possible simply from the astronomical variety of possible proteins. We can see too, in principle, how the specificity probably arises—through the effect that a quite extended amino acid sequence can have on the detailed folding of the final protein molecule, and in particular on the configuration at the active centre. The folding of the enzyme protein, then, does at least two things. It converts a linear code into a subtle three-dimensional object, and it thus *concentrates information* distributed along a chain at a particular region: the extended sequence defines an active centre.

In a series of theoretical papers on layer-lattice silicates, Radoslovich (1962, 1963) and Radoslovich & Norrish (1962) have stressed the importance of detailed packing and valence considerations with respect to substitutions in these structures. Van Olphen (1963, p. 78) has pointed out that the colloidal dimensions of many layer-lattice silicates are probably due to the creation of tensions in the lattices, caused by substitutions, giving rise to weak spots. In an electron diffraction study of calcium montmorillonite, Cowley & Goswani (1961) found that the sheets were appreciably bent within coherently diffracting regions of the specimen, a conclusion supported by electron microscopy. From an effect observed with parts of their specimen with organic molecules, these authors concluded tentatively that some rather bent crystal sheets selectively bound organic molecules, and that this resulted in straightening of the sheets. Whether or not the above are indications of a potential "tertiary structure" in clays, the following surface and edge effects of substitutions might be expected:

(1) The surface will become covered with a pattern of negative charges arising directly from the substitutions.

(2) A pattern of strains will be created due to substitutional size differences, giving rise to

(3) special regions where, through the accumulated effects of substitutional strains, lattice components are missing (e.g. a cation—giving a negative hole, or an $O^{2-}$ ion—giving a positive hole); or

(4) odd-sized sites where "foreign" atoms are incorporated.

(5) By specifically limiting the growth of the crystal (of van Olphen, 1963, p. 78), (2) might also give rise to special edges.

(6) To relieve strain the sheets might become bent in a complex way that depends on the substitution pattern. (In the two-layer clay haloysite, the sheets actually roll up into tubes—here probably due to consistent misfitting of a different kind—between the $A_1$ and $B_1$ oxygen layers (Fig. 8(a)).)

Industrial clays are usually "activated" with acid to enhance their catalytic and adsorptive properties. This treatment partly destroys the clay lattice, and the activity is probably due to surface features so introduced (Rvland *et al.*, 1960, pp. 14–15).

From the above considerations we might expect that a genetically controlled substitution pattern—which would represent a single and biologically selected isomer—could give rise to genetically controlled adsorptive and catalytic effects: in particular, that information could be concentrated at special active centres through the accumulated effects of substitutions that were spread over a relatively large area. (Compare the concentration of information in an enzyme protein.) Thus while initially the control functions of such a system would have to be very simple, relatively sophisticated and highly evolved forms would seem, in principle, to be possible.

The case for a clay as having been the primitive genetic material rests very largely, of course, on the earlier general theory leading to the idea of a genograph—that the primitive genetic information was held in *some* kind of crystal in the form of *some* kind of imperfection pattern. The more specific arguments can be summarized as follows:

(1) *Strictly genetic functions.* Clays can in principle hold large amounts of information in a particularly stable way: there is evidence that at least one layer-lattice silicate (mica) could replicate such information.

(2) *Function as a skeleton or membrane.* Clay particles can hold and concentrate organic and other molecules and ions (Bernal, 1951).

(3) *Control functions.* A substitutional genograph in a clay, could probably have particularly subtle adsorptive and catalytic effects.

(4) *Availability.* As pointed out by Bernal (1951), the involvement of clay in the origin of life (or processes leading to the origin of life) is suggested in part by its great abundance, and by its association with rivers— a point which will be considered in the next section.

From a practical point of view there are "disadvantages", in that clay synthesis under normal conditions is slow and not well understood, and also that the details of clay structures are still far from resolved. On the other hand, while it would be very difficult to synthesize a specifically functioning substitutional pattern, it is implicit in the above ideas that such a structure could be "bred" (see notes on p. 70).

### The specific (genographic) theory of the origin of life

(Compare the general theory as stated on p. 66.)

"The primitive genograph was a pattern of substitutions in a colloidal layer-lattice silicate."

### Comment

*Silicon and carbon.* Iler (1955, p. 277) has suggested that silicates may have been essential in early living things, and has pointed out that certain

ancient forms of life employ silica in their structure. The idea that silicon could replace carbon's key role in organisms has been opposed by Oparin (1957, p. 107), and by Wald (1964): according to the above specific theory, however, silicon was, in a sense, the key element in primitive organisms; not as a substitute for carbon, but in fulfilling a special role for which carbon would have been quite unsuited. There is, however, a similarity between the modern and the proposed primitive control mechanisms: nucleic acids and proteins are molecules that are also "crystals" (Calvin, 1961), while silicates are crystals that are also molecules. In both, the whole orchestra of physico-chemical forces—from Born repulsion to covalent bonding—contributes to the final configuration. According to the view being proposed here, the difference is that the primitive genetic machinery could form spontaneously, being a straightforward crystal, and act directly, the information being held on a surface rather than in a sequence; while the modern genetic machine requires a very special environment in which to work, although its indirect mode of action greatly increases the range of its effects.

### 4. Speculative Outline of Primary and Secondary Origins of Life

From the general theory of the origin of life described in section 2, it would follow that there would not be a unique origin of primitive life, but separate "origins" occurring continuously—and indeed still occurring—involving different kinds of crystals in different kinds of environments. In the special theory we considered only one kind of crystal: we will consider now only one imaginary evolution in one of a number of possible environments to illustrate the more general ideas.

Let us suppose (cf. Bernal, 1951) that there was a division between land and sea on the primitive earth, and that, as under present conditions, there was a water cycle producing dilute solutions of silica, and other precursors of clays, through the weathering of rocks. We suppose also, on the other hand, that in the atmosphere and elsewhere relatively simple organic substances were present in reasonable quantities through the action of various agencies in a reducing environment. This latter supposition is based on much theoretical and experimental evidence and has been very fully discussed (e.g. Calvin, 1961; Calvin & Calvin, 1964; Fox, 1963; Oparin, 1957; Bernal, 1951; Miller, 1953; Miller & Urey, 1959; Harada & Fox, 1964; Ponnamperuma et al., 1963; Ponnamperuma et al., 1964).

Surface and underground water courses would be formed in such an environment, containing molecules like amines, aminoacids, aldehydes, heterocyclic bases, random or ordered—but not *organized*—polymeric material, as well as clay-forming minerals. The protracted time scale of

geological events would ensure that an overall constancy was maintained for long periods, while annual, diurnal and—on coastlines—lunar cycles would provide limited periodic variations in the relative concentrations of the dissolved substances, in temperature, and in other factors. (If we think of the primitive earth as a machine with a kind of metabolism, in that it maintained such cyclic flows of materials, then we can think of the primitive organisms as *viruses* adapted to the earth as host—making use not only of matter and energy provided by the earth, but also of its "metabolism". This should become clearer later.) Clays would periodically crystallize from these dilute flowing solutions (clays can be formed under simple dilute flow conditions in the laboratory (Henin, 1956) and give rise to genographs. We suppose that in small clay particles, random genographic variations would produce differences in the "spectrum" of adsorbed molecules, which in turn would give a distribution of physico-chemical properties in the composite particles.

A genograph would have survival value if through its adsorptive or other effects it:

(1) tended to keep the clay particle holding it within the clay-synthesizing environment, so that it could act as template for further clay synthesis, or

(2) improved the physico-chemical stability of the clay particle holding it, or

(3) favoured replicative efficiency, i.e. clay crystallization and layer cleavage.

Imagine, for example, that clay-forming solutions are flowing through a bed of particles—e.g. sand—constituting a clay synthesizing environment. This could produce a "chromatographic" effect on the colloidal particles being formed, the rate at which they move depending on the nature of their adsorbed molecules. The population would become more and more slow-moving through evolution due to (1) above. But more subtle effects could also be expected. If the particles were too self-adhesive they would tend to clog the channels in which they were forming, and so stop further synthesis. The "problem" would be to stay in the sandbed without stopping the local flow of "food". This might be achieved by the particles acquiring an anchor from the environment—e.g. some organic polymer—with which they could adhere to the sand particles but not to each other (Fig. 13); or perhaps more simply through their preferential adsorption of organic molecules with which they could form a gel (Fig. 14) that would hold the genetic crystallites in place while still allowing the clay forming solutions to diffuse past them. By exerting some measure of selection on the diffusing molecules and ions in the vicinity of the clay particles, this gel could in its turn affect factor (2) above, which could be important, particularly under recurrent
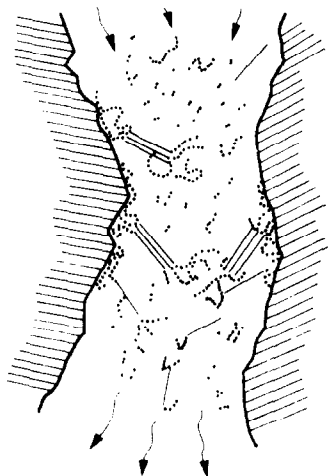
FIG. 13. A genetically controlled ability to form strong attachments to certain environmental polymers might anchor clay particles within the flow environment that had favoured their synthesis. If binding were at the edges of the crystallites, it would be less likely to interfere with further (replicative) clay synthesis.
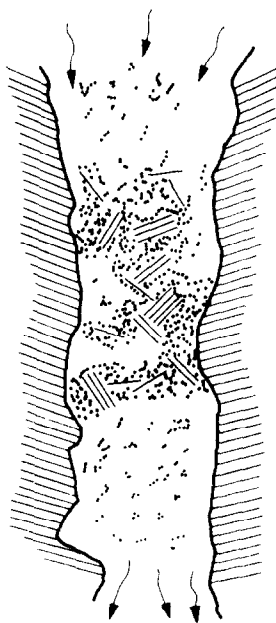


FIG. 14. Through selective adsorption of organic molecules in the environment, a group of genetically related clay crystallites might form a protective gel which could act as an anchor, as a buffer against periodic adverse environmental conditions, and as a favourable immediate environment for clay synthesis.

"difficult" conditions that tended to dissolve or otherwise "kill" clay particles. Factor (3) could similarly be affected, since crystallization processes may be inhibited or favoured by impurities present in the surrounding solution (Gilman, 1963). In such ways the formation of subtly adapted gels could favour survival of the clay particles within them—and natural selection would thus tend to produce such adapted systems.
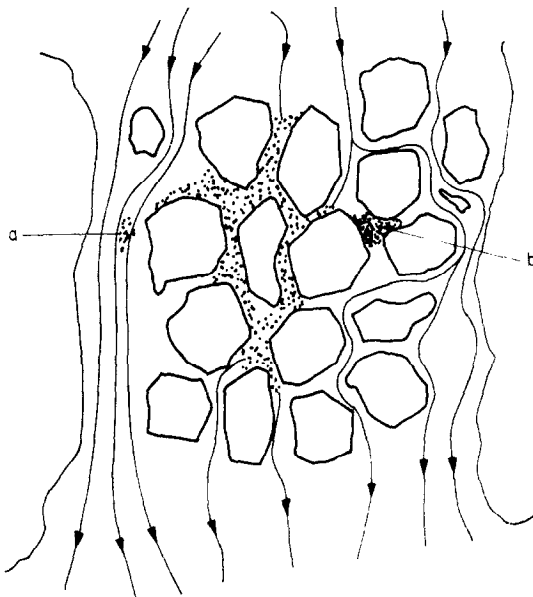


Fig. 15. A hypothetical multi-gene primitive organism in a stream flowing through a sand-bed (Fig. 14) would grow, particularly upstream, if the gel was loose enough for the solution to flow and diffuse through it. (a) Part of the growing organo-clay gel breaking off on encountering a region of rapid flow. (b) A less successful organism stopping local flow of solutions ("food") by forming too compact a gel.

Figure 15 illustrates the development of such an organism. The organo-clay gel would spread through the channels between the sand grains. A successful mutation in part of the gel net would increase the growth rate in that region. Growth would also be favoured in directions rich in "foods", and would die out in directions that no longer favoured clay synthesis, or where the required organic components were missing. In this sense the organism —or perhaps it should be called a colony of organisms—would move towards local regions where conditions for growth were optimal. This would tend to be up-stream where the concentration of "foods" was highest. Another effect that could spread successful genographs—down-stream—

could be if part of the growing net were to encounter a region of rapid flow so that pieces of the organo-clay were broken off to re-establish themselves elsewhere. A process of vegetative reproduction would then be complete.

We imagine this organism growing and evolving, then, by a combination of clay synthesis and an increasingly specific accretion of organic molecules, etc., so as to form a gel rather like the complex co-acervate droplets described by Oparin (1957). Unlike Oparin's first organisms, however, such an organo-clay complex could start to evolve through natural selection before any consistent chemical reactions were established. The only *essential* dynamic feature of an organism itself is in its genetic replication: the other dynamic features can be in the environment. We imagine that these very early organisms were largely "run by the environment" (compare again viruses), their only intrinsic dynamic properties having been the physico-chemical processes of crystallization and crystal cleavage. We suppose that abilities to bring about chemical reactions developed later—through the selection of genographs that had suitable catalytic centres. Such abilities could clearly have been advantageous for the primitive genes, by giving them a greater variety of molecules out of which to form their protective immediate environments (i.e. their "bodies", cf. p. 76), and thus making them more independent of their general environment. Evolution would tend to produce more and more active organisms, able to create their own internal flows, and so able to survive under less dynamic external conditions.

### Genetic metamorphosis and the secondary origin of life

It has been the main purpose of this paper to consider what we have called the *primary* origin of life: it has been the fundamental attitude that this origin was a strictly physico-chemical event that can be understood in terms of the inherent self-organizing properties of matter; that it can be investigated through studying the physico-chemical properties of commonplace materials; that it is in this sense quite accessible. The molecular mechanisms by which modern organisms function are also, and increasingly, accessible. By contrast, what we have called the secondary origin of life is still very inaccessible, being separated by two long evolution processes from physical chemistry on the one hand and molecular biology on the other. The general arguments for *some* kind of genetic metamorphosis have been considered in the early part of this paper; it remains to indicate that the specific theory is thinkable: that a "take-over" of genetic control, from inorganic clay crystals by organic macromolecules, was, at least in principle, possible. We will consider a hypothetical clay → pseudo-nucleic acid metamorphosis.

The key phenomenon is envisaged as being the formation of two-dimensional crystals of organic molecules between the silicate layers in clays (cf. p. 68).

In evolved genetic clays one can envisage a specific complex array of organic compounds forming a kind of organized, mixed, two-dimensional crystal, in which the arrangement of the organic compounds reflects the substitutional information in the silicate layers.† Reactions occurring in such an array containing suitable monomers could give rise to polymers with a genetically controlled configuration, out of which secondary control structures, membranes and other cell structures could be formed. We could call such a polymer a "pseudoprotein" without implying that it was necessarily a polyaminoacid. There are indeed many possibilities here. An example of a particularly simple kind of informational macromolecule could in principle be produced by the polymerization of formaldehyde, to give poly-CHOH, through the aldol-type reaction that has often been suggested as a possible abiogenic source of sugars (see Oparin, 1957, p. 198). Imagine, for example, a close packed interlaminer array of cations and formaldehyde molecules in which the latter are arranged in rows—corresponding, perhaps, to rows of substitutions in mica-like domains in the silicate. Now suppose that the following rule applies: where a row is straight the resulting polymer is isotactic (i.e. all the OH groups are on the same side of the carbon chain) but that where a row changes direction—e.g. at a mica-like domain boundary—there is a reversal of tacticity in the resulting polymer: then, genetic information held as a (domain) pattern in the silicate could be translated into a "pattern of tacticity" in the polymer. Information of this kind might well determine the way in which such a pseudoprotein would fold up in free solution. Sugars (small oligomers) with a genetically controlled configuration could also be produced by this mechanism.

Another general possibility would be the formation of informational co-polymers through the reaction of two or more kinds of specifically orientated monomers within an interlaminar complex.

We can also consider two variants of the idea that is often proposed that a polymer might be able to replicate through crystallization of suitable monomers on pre-existing polymer. Such a process can be envisaged within a clay interlaminar region, starting with the adsorption of a preformed polymer

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† In the systems studied by Weiss (1963), and referred to on p. 75, the layer-lattice silicate itself determines the *proportion* of alkylammonium ions in the closely packed, mixed, interlayer complex. (There are two organic molecules per $(Al, Si)_4O_{10}$ unit of which from 0·25 (hectorite) to 0·92 (muscovite) are the organic cations. This is so for a range of chain lengths and for different kinds of second components.) It remains to be shown whether the relative *positions* of the organic components are also determined by the silicate.

which then determines the orientation of adjacent monomeric components during the formation of a two-dimensional crystal. Figure 16(a) illustrates a possible arrangement for the replication of a polymer, such as poly-CHOH, where the information is "written" in its tacticity. A complementary duplication process, operating through size pairing, can be considered for an
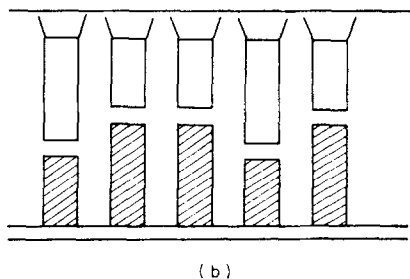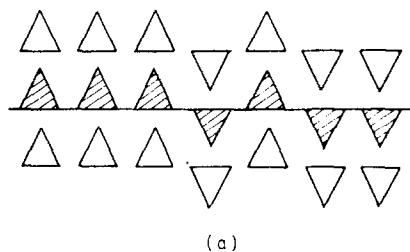


( a )



( b )

FIG. 16. It is suggested that the restricted interlaminer regions of clays provided "molecular clamps" that assisted in the replication of the first organic genes. (a) The complex tacticity of an informational homopolymer determines the orientation of adjacent monomers (leading to replication of tacticity on subsequent polymerization) or (b) Information in a binary co-polymer is transferred through the formation of a nucleic acid-like "ladder".

informational co-polymer (Fig. 16(b)). We suppose that gradually more and more of the genetic information would be stored within these polymers themselves, the function of the clay gradually changing from that of a template to a more passive role as a "clamp".

It would seem that the first organic gene to appear would have had to act directly within the organisms, i.e. perform both the functions of nucleic acids and proteins, although its structure may have been different from either—e.g. poly-CHOH. It may, on the other hand, have been something like protein. Some globular proteins take up a $\beta$-configuration in forming clay interlaminar complexes (Talibudeen, 1955). It is possible that a protein-like organic macromolecule might take up a linear "replicating" configura-

tion, as in Fig. 16, when inside a clay "clamp", but fold into a functional information-dependent tertiary structure when in the "cytoplasm".† If this was so, then the hereditary machine would still have depended on clay even after the more strictly genetic functions had been taken over: it would only have been when two separate molecules had appeared, with some kind of organic go-between mechanism, i.e. a pseudo-DNA/RNA/ protein system, that clay could have finally disappeared from the control machinery of organisms.

We can think of a genetic metamorphosis as having occurred then, in three broadly overlapping stages, with long periods in which mechanisms corresponding to different levels of development were operating side by side in the same organisms.

(1) Transfer of control function from the clay surfaces themselves to organic macromolecules formed on the clay surfaces, i.e. the formation of pseudo-proteins.

(2) Transfer of replicative function from clay to replicating pseudo-proteins.

(3) Division of replicative and control functions between two different kinds of molecules, i.e. the appearance of something approaching the modern system with the concomitant disappearance of clay from the genetic machinery.

Certainly such a description leaves many questions to be answered, and it may well be wrong in many important respects. But whatever the exact stages in the process, the appearance of a sophisticated organic hereditary mechanism is more understandable *within a working (primitive) organism*, since then partial functions can be selected for: even a very simple organic gene could confer survival value on an organism that held it, although such a gene might have no significance outside the organism. It is much easier to add to a going concern than to start *de novo*.

Finally, taking a very broad view of the analogy referred to previously, we might say that the very first organism was the primitive earth itself; that primitive (clay-based) organisms were initially viruses, with the earth as host, which developed more independence through the establishment of a metabolism in their associated organic structures, and that the subsequently forming organic genes started in their turn as symbiotic viruses within these clay-organisms. Thus, through perhaps many such hierarchical stages, the modern hereditary mechanism may have evolved.

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† This was perhaps the original significance of the $\beta$- and $\alpha$-configurations of proteins. Haldane (1965) suggested that RNA preceded DNA, and asked the question, "Can protein be replicated without RNA?"

## 5. Notes

### THE PROSPECT OF FORMING ORGANISMS ARTIFICIALLY

According to the idea of spontaneous generation ((1), p. 54), "artificial life" is a feasible proposition (because systems that are far more "improbable" than the first organisms, e.g. insulin molecules, have been made). According to the idea of "chance and time" ((2), p. 54) such a project might require an infinite induction period. The idea of abiogenic evolution ((3), p. 54) also suggests that an indefinite time-lag might be involved. From the idea of genetic metamorphosis ((2), p. 58) that has been pursued here, it follows, as with (1), that artificial organisms could be made, but that these would be primitive organisms. As with (3) an indefinite evolution period would be required for the appearance of organisms using only organic genetic machinery. But even primitive organisms could be extremely useful, and the possibility of "breeding" specific industrial catalysts is inherent in the genographic theory of the origin of life. Such could be produced by artificial selection even before fully self-selecting systems appeared.

One would generate primitive organisms by applying a biological technique to a physico-chemical system: by setting up a suitable cyclical flow "environment" and waiting for the organisms to appear. In general the conditions would be such as just permitted the crystallization of some potential genetic material, and where a crystallite would only avoid subsequent destruction if it were able to adsorb specifically some kind of molecule or molecules in the surrounding solutions (section 4). When simple adsorption organisms had been established, the external conditions could be made "harsher" and more complex so that a mutant genograph would be strongly selected for more specialized and complex functions.

### VARIETY OF LIFE FORMS

According to the view that has been pursued here, the significance of the formation of "biochemical" molecules under assumed primitive earth conditions (referred to under (3), p. 55) is not that such amino acids, purines, etc., were essential for the origin of life, but that because they were present under primitive conditions, they were incorporated into the primitive organisms that were the ancestors of the modern form. This implies that on other planets—where the "spectrum" of abiogenically formed organic molecules would not necessarily have been the same as on the primitive earth —any organic life form that might be present would not necessarily be expected to involve the same fundamental polymers. The possibility that life on other worlds will be found to be "primitive" is also, of course, implied.

## PRIMITIVE GENES ON THE MODERN EARTH

As already indicated there is a rather trivial sense in which almost any crystal is a primitive gene. Very limited primitive evolutions might also be expected under natural conditions. But a more interesting possibility is that vestiges of primitive genetic machinery may be present in "ancient" organisms, particularly those closely associated with silica—e.g. lichens or diatoms.

Also the possibility of parasitic primitive organisms is implied. Such "inorganic viruses" might form spontaneously, and evolve, within the cyclical flow environment of another organism. Alternatively, a replicating catalytic centre that brings about a reaction that interferes with normal metabolism—a malign genograph—might appear in traces of normally harmless solid inorganic material within an organism. Silicon might not necessarily be involved, and even if involved in the second sense above, there might be no great disturbance in the normal silicon content, but the (admittedly low) normal silicon content of man might turn out to be of significance as Iler (1955, pp. 293–296) suggests, and the association of cancer with intake of asbestos dusts (Wagner, 1962) and with soil environment (Stocks, 1961) is of particular interest in this connection.

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