

Table 1. The effect of various substances on the absorption of gamma radiation from I^{125} by the concentric tubes technique. The thickness of the annular space was 2.0 mm.

Substance	Density (g/ml)	Transmission (%)
Air (control)		100
Water	1.00	94
Potassium tartrate	1.40	64
Chloroform	1.47	34
Carbon tetrachloride	1.60	30
Cesium chloride	1.60	12

sis, alcohol-precipitation, or chromatography.

Although corrections for this absorption may be applied, or the use of salts with nuclei of high atomic number avoided, an awareness of this absorption phenomenon may be of value in the choice of the radioactive isotope to be employed.

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Condensation Model Producing Crystalline or Amorphous Tetrahedral Networks

Abstract. Wire models of tetrahedral networks have been built up by a process representing condensation from a fluid, which yields amorphous structures unless a nucleus of crystalline structure is present initially. A random tetrahedral network of 300 units has been constructed for direct measurement of coordinates; the construction process is uniquely defined so that it may be possible to synthesize larger networks by use of a computer.

In constructing experimental networks of tetrahedra linked at all vertices (to serve as models of tetrahedrally coordinated atomic arrangements in substances such as SiO_2 and H_2O), an attempt was made to codify the con-

struction procedures so that ultimately the construction of the network may be done by a computer. The object of the work is to realize at last the "random network" that has been discussed as a rather vague abstraction ever since Zachariassen (1) and Warren (2) first introduced the idea.

The random network is conceived as a structure in which the relation between adjacent units is defined rather rigidly by the coordination and any limitations on bending or rotation of bonds, but the relations between more distant units are variable, depending on the variations of bending and rotation within permitted limits. Because a specific model was not available, Warren (2) approximated the distribution of distances for nearest and second nearest tetrahedra by analytically defined distribution functions that ignore any influence of the remainder of the network on the orientation of these units. More detailed random network models may yield closer correlations with experimental data and thereby permit more critical examination of the postulates on which the models are based.

Regular structures (3, 4) have also been postulated which embody the same principles of bonding as the random network. Experience with the present models suggests that these regular structures are rare, although they may be important in special nucleation processes. The geometrical and topological statistics of the regular models are easily calculated, but serve only as rather unsatisfactory approximations to those of the random network. A specific random network model must be large, however, in order that its statistics may represent an adequate sample of an infinite population.

The wire tetrapod units devised by Tilton (3) are used, primarily because the great emptiness of the resulting model makes it possible to determine the coordinates of each unit, but also because the units are so conveniently joined (Fig. 1).

The ends of the four 5-cm legs of each tetrapod, made of wire 1.5 mm in diameter, correspond to the vertices of the coordination tetrahedron; their junction corresponds to its center. Joining two tetrapods by a length of suitable elastomer tubing forms a bond that tends to remain straight but permits free rotation of each tetrapod about the bonded leg. The addition of new tetrapods and new bonds between previous ones can be continued, with the

permitted rotations and only moderate bending of bonds, to produce amorphous structures or any regular network based on tetrahedral coordination.

Adding a unit to the model involves three stages of decisions: the site at which the new unit is attached, the positions to which its other three legs are rotated, and the formation of any bonds to these other three legs. The following set of rules was found sufficient to generate an amorphous network of 300 tetrahedra with no unshared vertices except those normally present at the surface, and with no apparent limit to further additions.

1) The new tetrapod is attached to the most highly bonded of the earlier ones that are still incompletely bonded.

2) Tetrapods are numbered serially in the order of addition to the model. If the choice of a site under rule 1 is not unique, preference is given to the tetrapod of lowest serial number.

3) The legs of each new tetrapod (and their projecting ends, hereafter called the vertices) are numbered 1-4, with 1-3 forming a right-handed vector system. The first bond to the model uses leg number 1. If the choice of a site under rule 2 is not unique, preference is given to the leg of lowest number.

4) When a new tetrapod is attached, the bond is initially straight, and the tetrapod is oriented by rotation about its bonded leg to the position that brings its lowest-numbered unbonded vertex closest to an unbonded vertex of another tetrapod.

5) After the attachment of each new tetrapod, one or more additional bonds may be possible without excessive deformation of the model (that is, bending any existing bond more than about 20 deg.). These additional bonds are formed one at a time, the preference being given in each case to the pairs of vertices closest together.

6) In the course of bonding under rules 4 and 5 it sometimes happens that two possible sequences of bonding are approximately equally acceptable, one of which leads to the inclusion of an unbonded vertex within a cage in the model. The choice is made to avoid including the unbonded interior vertex. This choice has always been found reasonable with tetrahedral networks, but other types of networks may quite possibly lead inescapably to the inclusion of unbonded vertices within the model.

Rules 1 to 3 are purely topological,

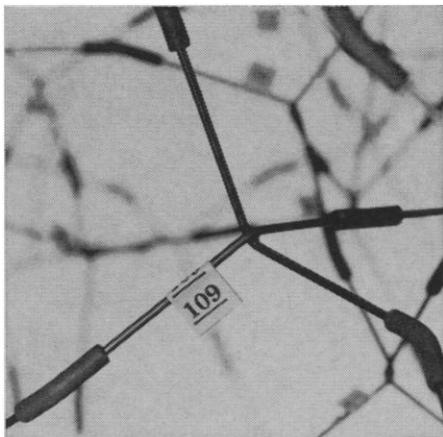


Fig. 1. A wire tetrapod unit as it appears in the random network.

and can be applied rigorously even in constructing a crude mechanical model. The remaining rules involve distances, which can be realized only approximately, and judgments concerning "excessive bending" of bonds which are entirely subjective. Therefore it cannot be said that the tetrapod models are always unique, which would be true of computer-synthesized networks. Computer synthesis in direct analogy to the construction of the tetrapod models is possible in principle, with appropriate mathematical representation of the network's adjustment to local deformation and the criterion of "excessive deformation" in rule 5. It is expected, however, that the most desirable computer synthesis will be a mathematically simpler one not completely analogous to the building of tetrapod models.

Rule 1 may be interpreted as a state-

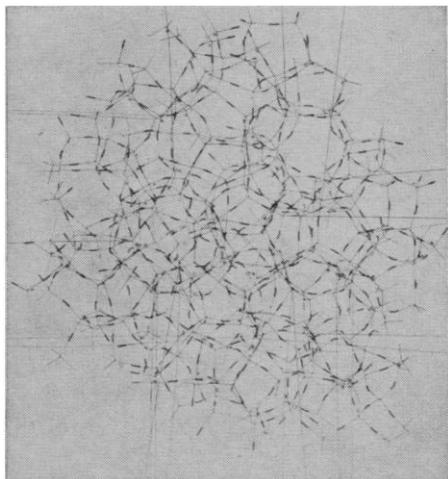


Fig. 2. The random network of 300 tetrapods, with the supporting wires used to hold it rigidly for measurement of coordinates.

ment that the condensation process involves a sufficiently low supersaturation and high rate of surface diffusion that each new unit attaches at the site contributing least to surface energy. If the order of preference in rule 1 is reversed, a loose, highly branched structure results which may have its analogue in the fluffy amorphous solids produced by certain condensation processes.

Rule 2 may be considered a statement of the fact that the probability of arrival of a new molecule at a given site increases with the length of time the site is available. Rule 6 may be considered a statement that the bond energy and the temperature are high enough for rearrangements to eliminate all interior defects in bonding.

These rules, followed from the beginning, lead to a compact noncrystalline structure (Fig. 2). In the earlier stages it resembles the regular structures proposed by Tilton (3) and Pauling (4). As the model grows beyond the size of Tilton's "13-unit vitron" (a pentagonal-dodecahedral cage surrounded by a complete layer of similar cages) all further growth is in what may truly be called a random network.

In order to avoid a possible statistical bias a portion of the model containing the regular nucleus could be removed or a nucleus that is not regular could be used at the start. This apparently is the first synthesis of a fully random tetrahedral network, although the tetrahedral network model constructed by Bernal (5) may be experimentally indistinguishable. Bernal's model has greater local regularity in that each tetrahedron belongs to at least one five-membered ring having the specific keatite (6) configuration.

If a six-membered ring in the chair form is present as an initial nucleus, the same rules lead equally unequivocally to the ideal high-cristobalite structure (7) consisting entirely of such rings. If the nucleus is a six-membered ring in the boat form, a cage of the high-tridymite (8) structure is produced, but thereafter some five-membered rings develop with inevitable loss of the structural regularity.

The most glaring deficiency of the condensation process under these rules is that sites offering a possibility of forming several bonds at once are given no preference, and therefore the completion of rings and cages is slower than it should be. Correcting this deficiency would, for instance, cause the six-membered ring nucleus in the boat

form to produce a perfect tridymite structure. Another improvement in verisimilitude would be to add the tetrapods in groups as well as singly. These changes would increase the complexity of tests or "false starts" during the construction process, at the cost of computational efficiency as is often the case when a Monte Carlo calculation is made more realistic. The construction of satisfactory random network models does not seem to demand such changes at present.

The procedures for efficient, precise determination of the coordinates, and the data on statistical topology and geometry of the random tetrahedral network model containing 300 units, will be published elsewhere.

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Cellulose Acetate Membranes: Electron Microscopy of Structure

Abstract. Electron photomicrographs of cellulose acetate membranes used in the reverse osmosis process of water desalination reveal a dense surface layer with a porous substructure. The high rate of transmission for water can be correlated with the thickness of the dense layer on the air-dried surface of the membrane.

Cellulose acetate films have been found effective in the desalination of sea-water brines by the so-called reverse osmosis process (1). Recent advances in the technology of film preparation have led to greatly improved water transmission rates (2). Preliminary investigation of film structure by optical microscopy has shown that the details of the structure are not apparent