

# ON THE PHYSICAL ORIGIN OF BIOLOGICAL HANDEDNESS

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**Abstract.** In the racemic conglomerate crystallization of over 1000 samples of D,L-sodium-ammonium tartrate the effect of  $^{32}\text{P}$  beta irradiation on the weight, optical activity, and crystallite size was measured. Both weight and optical activity showed a statistical dependence on the intensity of beta irradiation. The crystallite size is also affected by the presence of  $^{32}\text{P}$ . Asymmetric crystals are suggested to have been potential mediators between asymmetric parity violating forces and molecular asymmetry so that stereo-selective prebiotic chemical reactions involving crystals need not be considered 'chance' processes.

No measurable difference in the energy content of optical isomers was found. An upper limit for the direct contribution of weak interactions to electromagnetic ones has been calculated. The mechanism of stereoselective crystal seeding by beta particles is discussed.

## Introduction: Chance versus Necessity in the Origin of Chirality

One handedness at the molecular level is a basic property of our living world. Living systems are composed of L-amino acids and D-sugars which when polymerized form right handed helical structures in proteins and nucleic acids, respectively. Optically pure polymers (e.g., polyamino acids) are more stable than the racemic ones (Blout and Idelson, 1956; Harada, 1970). These secondary helical arrangements play important roles in many fundamental molecular processes in biology. This is one of the reasons why asymmetry offers a selective advantage during chemical evolution over symmetric, racemic systems as the level of organization increases.

Contrary to the findings in biochemistry (Ulbricht, 1962), the simulation experiments of prebiological chemical events yield racemic mixture of enantiomers (Wald, 1957; Miller, 1973). *How* then could an asymmetric life emerge from symmetric starting materials?

The origin of asymmetry can be approached in two different ways: (a) asymmetry has evolved by a chance process and (b) asymmetry is produced by some known physical agent.

### ASYMMETRY BY CHANCE PROCESSES

Chance process means a non-repetitive single event caused by apparently unknown forces happening only once – or very few times – within a relatively narrow time interval in the history of earth.

Repeatability is considered a crucial requirement in proving any statement in science, and the same is true for the evolution of life. Involving chance means that selection

between left and right, L and D forms, would be a lone exception (Elias, 1972). As a result this hypothesis is not experimentally testable.

It has been shown (Secor, 1963; Pincock *et al.* 1974) that in any individual trial partial crystallization of racemates may result in self-propagating separation of enantiomers. In fact, racemates rarely supply exactly equal amounts of L and D isomers upon crystallization (Thiemann and Darge, 1974). Nevertheless, as the probability of right and left handed seeding is the same, after a sufficient number of crystallizations one expects the mean optical activity to tend to zero. This has been verified experimentally (Pincock *et al.*, 1971; Kovacs and Garay, 1975; Kovacs, 1978).

The evolutionary significance is that because of statistical fluctuation no optical activity will develop if the number of events permits statistical description of the system.

#### ASYMMETRY PRODUCED BY KNOWN PHYSICAL AGENTS

An asymmetric physical source of molecular handedness has to meet the following criteria:

- it has to represent a bias in the same direction for an extended period of time
- it has to occur everywhere on the Earth to be able to interfere in the start of evolution at any appropriate place
- it has to be linked to some of the available energy sources present on the surface of primordial Earth.

So far two suitable routes have been proposed.

*Circularly polarized light.* Kuhn and Braun (1929) and later several other authors have shown how circularly polarized light can participate in stereoselective synthesis (Tenney and Ackerman, 1945; Buchardt, 1974) and decomposition (Terent'ev and Klabunovskii, 1960; Kagan *et al.*, 1974). Byk (1904) suggested first on theoretical grounds that the circularly polarized components in the reflected sunlight should be distributed differently.

The only flaw from the evolutionary point of view is that in spite of these well established facts the sunlight contains only a very small fraction of circularly polarized light (Kawata, 1978).

*Weak interactions.* Parity violation in weak interactions (Lee and Yang, 1956) offers another possibility for absolute asymmetric prebiotic processes (Vester, 1957). Because of the minuteness of the effect, however, both positive and negative results have been gathered (for a recent critical review of the related experiments see Keszthelyi, 1977b).

#### Possible Biological Consequences of Parity Violation

In principle there are two mechanisms which can carry the elementary particle asymmetry over to the molecular level.

*Vester-Ulbricht process.* In this process the role of weak interactions (Vester and

Ulbricht, 1962) is simply to produce the spin-polarized particles. The polarization takes place during the emission of the beta particles. In the surrounding material the beta particles lose their velocity but may keep a substantial part of their polarization. The interaction of the slow polarized electrons with optical isomers is governed by electromagnetic forces, and spin-orbit coupling between beta electrons and valence electrons of the molecules can bring about asymmetric radiolysis (Garay and Hraskó, 1975; Keszthelyi, 1977b).

*Yamagata process.* Due to the mixing of weak interactions with electromagnetic ones an asymmetric contribution is present in intramolecular electromagnetic processes too. It will result in an energetic nonequivalence of optical isomers. Yamagata (1966) proposed a mechanism for the amplification of this small difference during polymerization of biomolecules. A theoretical treatment of this idea has been given recently by Letokhov (1975).

In the crystallization experiments to be discussed here both aspects were studied experimentally. In addition, the parameters of crystallization taken as a function of radioactive concentration was considered crucially important to prove any effect of beta particles.

### Asymmetric Crystals

Several different possible roles of crystals in chemical evolution have been recognized.

From the point of view of the origin of life's preference of one optical isomer to its antipode, the existence of separable mixtures of asymmetric crystals is often regarded as essential (Northrop, 1957; Harrison, 1974). Several compounds are known to form racemic conglomerates, i.e., a mixture containing identical amounts of individual crystals of each antipode is deposited from the supersaturated solution of the racemate. Once the ratio of L/D crystal seeds is shifted from 1 for any reason, the absolute difference will increase as crystallization goes on due to the autocatalytic nature of the process (Calvin, 1969).

In his famous crystallization work Pasteur (1848) took advantage of the same phenomenon and discovered optical isomerism by separating the two isomers of sodium-ammonium tartrate. He felt the almost complete stereoselectivity in living beings was a consequence of some 'cosmic asymmetry'. Since then the hypothetical cosmic asymmetry was discovered as parity violation in weak interactions and stimulated active research. In this study Pasteur's original compound was used to investigate the modern version of his favorite idea.

### Materials and Methods

D,L-tartaric acid (Reanal) was recrystallized several times from water until it was optically inactive. Pure tartaric acid was converted to its sodium-ammonium salt and recrystallized again. Enantiomers of mandelic acid (Norse), phenylalanine (Merck) and leucine (Reanal) were used without further purification.

All optical activity measurements were done on a JASCO 40c dichrograph.

The method of crystallization of D,L-sodium-ammonium tartrate has been published in detail elsewhere (Kovacs and Garay, 1975; Kovacs, 1978). In the pilot study 63  $^{32}\text{P}$ -irradiated probes were crystallized together with 63 controls. In this study 900 independent crystallizations were carried out at three isotope concentrations. In each run 30 irradiated and 30 control samples were placed in one desiccator. The results of five experiments at each isotope concentration were treated together. In a few cases the sample formed a highly viscous, glasslike material instead of crystals. These were discarded.

## Results

### BETA PARTICLES INTERACT WITH DISSOLVED TARTRATES: WEIGHT AND CRYSTAL SIZE MEASUREMENTS

The distributions of dry weight of the crystalline phase were determined. At the low level of radioactivity no effect of  $^{32}\text{P}$  betas on the weights of crystalline material was observed. An increase of isotope concentration, however, was accompanied by a pronounced difference between irradiated and control series (Figure 1, details were given in Kovacs, 1977b). The weight in the  $^{32}\text{P}$  containing probes shifted towards higher values (Table I). Within the region studied, the weight increase was proportional to the logarithm of the radioactive agent concentration.

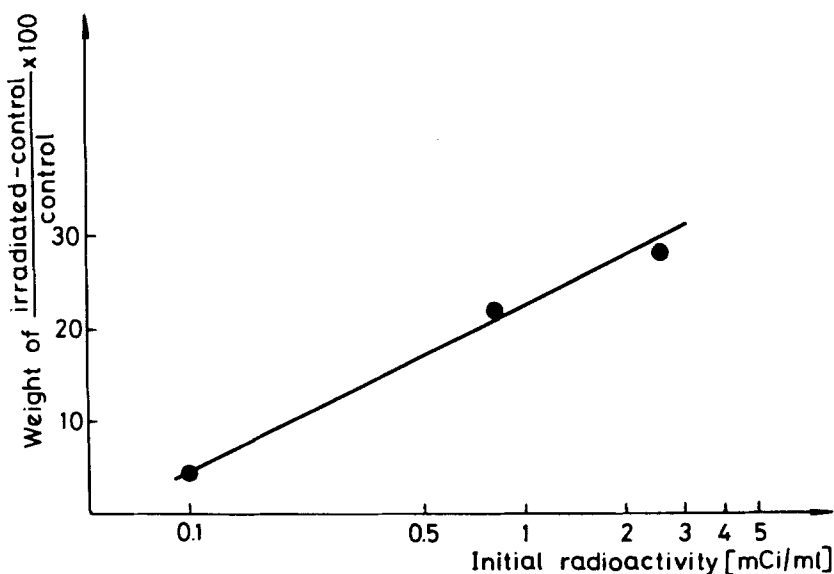


Fig. 1. Semilogarithmic plot of relative weight differences between control and  $^{32}\text{P}$  irradiated crystallizations as function of initial radioactivity.

TABLE I

The measured parameters of D,L-sodium-ammonium tartrate crystallization in the presence and absence of  $^{32}\text{P}$  beta irradiation

Radioactivity (mCi/sample)	Weight (g)	Optical activity <sup>a</sup> (deg cm <sup>2</sup> decimole <sup>-1</sup> )
5.0	0.333 ± 0.018	+16.14 ± 4.60
0	0.256 ± 0.017	-0.10 ± 4.21
1.6	0.231 ± 0.017	+12.62 ± 4.43
0	0.189 ± 0.020	-0.08 ± 3.66
0.1	0.202 ± 0.016	+2.43 ± 2.15
0	0.194 ± 0.015	+0.47 ± 1.74

<sup>a</sup>at 215 nm; for D-sodium-ammonium tartrate  $\langle \theta \rangle_{215} = -6850 \text{ deg cm}^2 \text{ decimole}^{-1}$ .

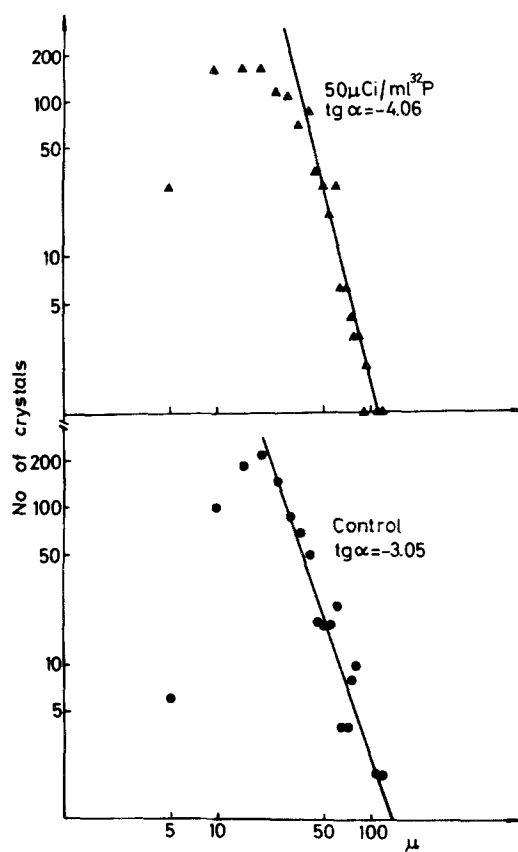


Fig. 2. Distribution of size of crystals from  $^{32}\text{P}$  irradiated and control probes.

In order to gain a better insight into the mechanism of tartrate crystallization the size distribution of deposited crystals was also studied. The wet crystals were smeared on microscopic plates and the lengths of their longest edges were measured under a light microscope. The distribution of lengths of 1000 irradiated and 1000 control crystallites are given in Figure 2. Anomalously, few small crystals were found. This is due partly to the resolution limit of the microscope and partly to the loss of many floating small crystals during removal of the mother liquid.

A straight line has been fitted to the data points above  $20 \mu$  using a least-square program. The difference in the slopes indicates an abundance of relatively smaller crystals in the irradiated samples. It is obvious from the above data, however, that no corresponding decrease in the average weight of crystalline phase took place. Consequently, a larger number of individual crystals formed in the presence of beta irradiation. In other words, beta particles increase the effective crystal seedings and lower the crystallites' average size.

#### THE INTERACTION IS STEREOSELECTIVE: OPTICAL ACTIVITY MEASUREMENTS

The crystalline phase recovered in the first 126 crystallizations (Kovacs and Garay, 1975) showed an average optical activity which corresponds to 0.01% optical purity. Significantly more L-sodium-ammonium tartrate crystals accumulated in the  $^{32}\text{P}$  containing probes.

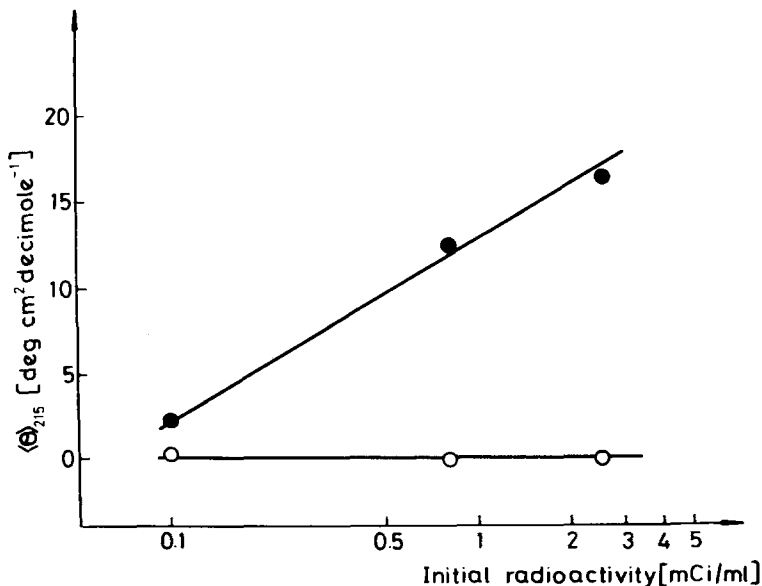


Fig. 3. Semilogarithmic plot of the observed average optical activities for control (—○—) and  $^{32}\text{P}$  irradiated (—●—) crystallizations as a function of initial radioactivity.

In the present work the dependence of induced optical activity on the amount of radioactivity was investigated. As with weight increase, optical activity showed a logarithmic relationship with the radioactive concentration (Figure 3). The numerical data are collected in Table I.

#### CRYSTALLIZATION IN THE PRESENCE OF OPTICALLY ACTIVE IMPURITIES

All other treatments being the same, only dissolved optically active impurities carried with the isotope solution could have accounted for the observed stereoselective effect. This possibility was checked by deliberately adding known amounts of optically active contaminants. The concentration of the entire dissolved material, including  $H_3PO_4$ , in the isotope solution that was added to the sample was less than  $0.005 \text{ mg ml}^{-1}$ .

The effect of four enantiomer pairs (sodium-ammonium tartrate, mandelic acid, phenylalanine, and leucine) was investigated and details have been reported (Kovacs, 1977a). The results relevant to the point of view of the present discussion are the following (see Table II):

(a) Optical activity is about 10 times more sensitive to the presence of impurities than weight increase.

(b) Nucleation of the tartrate isomer isomorphous with the added enantiomer is preferred. Similar observations were made by Kipping and Pope (1898), Ostromisslensky (1908), Secor (1963), Harada (1970), Pincock (1971), Thiemann (1974). The tartrate

TABLE II  
Effect of dissolved impurities on D,L-sodium-ammonium tartrate crystallization

Material	Lowest effective molar ratio $\times 10^2$		Optical purity <sup>a</sup> (%)
	Significant weight increase	Optical activity	
D/-/-sodium-ammonium tartrate	1.3	0.13	0.9
L/+/-sodium-ammonium tartrate	1.3	0.13	0.9
/-/-mandelic acid	0.4	0.04	1.6
/+/-mandelic acid	0.4	0.04	1.6
D-phenylalanine	1.3	0.40	0.8
L-phenylalanine	1.3	0.13	2.1
D-leucine		1.3	0.4
L-leucine		1.3	0.5

<sup>a</sup>at 1% impurity concentration.

optical activity versus contaminant concentration curves are mirror symmetric for opposite handed admixture pairs. This is in apparent contradiction with Thiemann's findings (1974). In the crystallization of D,L-asparagine Thiemann obtained a difference not only in the sign of induced optical activity but also in its magnitude upon adding extra L- or D-asparagine. He interpreted the deviation as strong evidence illustrating energy differences between left- and right-handed crystal lattices.

(c) 1–5 mg ml<sup>-1</sup> chiral contamination has to be present in the crystallizing racemate solution to cause significant optical activity of the crystalline phase. This figure is at least 10<sup>3</sup> times as high as the mass of dissolved materials carried with the isotope solution. Thus contamination is highly unlikely as an explanation of the stereoselective crystallization in the <sup>32</sup>P irradiated probes.

#### SEARCH FOR AN ENERGETIC DIFFERENCE BETWEEN TARTRATE ENANTIOMERS: NON-IRRADIATED CRYSTALLIZATIONS

Of 450 control experiments 440 yielded crystalline material. These controls served a double purpose: on the one hand they should have indicated any external factor responsible for differential crystallization of tartrate isomers, while on the other hand they allowed a thorough test of the suggested energy content nonequivalence due to direct contribution of weak interactions to electromagnetic ones (Yamagata, 1966). Thiemann and Wagener (1970) found stereoselective crystallization of D,L-sodium-ammonium tartrate in every one of 10 attempts. 440 samples seem to be enough to compensate for accidental statistical fluctuations which may explain their positive result.

The average optical activity of control probes was  $0.2 \pm 1$  deg cm<sup>2</sup> decimole<sup>-1</sup>, i.e. within experimental error no optical activity was obtained. This means that no outside stereoselective factor interfered in the crystallization. As controls and irradiated samples were treated together (except for addition of <sup>32</sup>P), the preferential crystallization of the 'unnatural' L-isomer is attributable solely to the presence of beta particles in those solutions.

In addition, no measurable evidence of any asymmetry effect due to the lattice energy difference between optical isomers was found. Theoretical considerations indicate (Rein, 1974) that such a difference must exist although its magnitude is certainly too small to be observed with our experimental techniques. Yet, an upper limit for the contribution of parity violating effects to intramolecular forces can be given. A Gaussian distribution has been fitted to the measured data (Figure 4) which has a half width of  $28 \pm 2$  deg cm<sup>2</sup> decimole<sup>-1</sup>. The asymmetry effect,  $\epsilon$ , can be calculated with the use of the following additional data (Keszthelyi, 1977a). The number of crystallites per test tube is approximately  $6.2 \times 10^4$ , the number of molecules that belong to one crystallite,  $n$ , is estimated to be  $5 \times 10^{16}$ , the relative shift of the distribution from zero optical activity is smaller than  $1.4 \times 10^{-4}$ .

According to Yamagata (1966):

$$2(P_L - P_D)/(P_L + P_D) \approx 2n\epsilon_1 \quad (1)$$



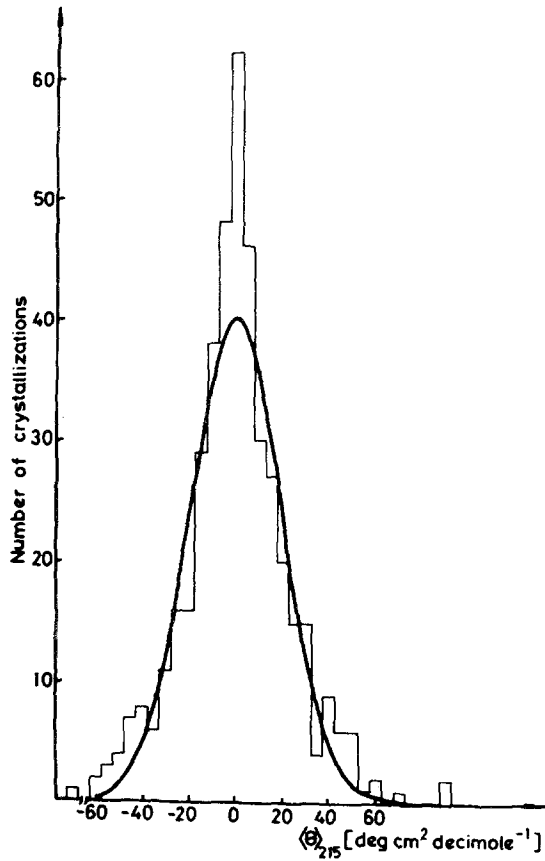


Fig. 4. Distribution of optical activities measured for the control crystallizations and the Gaussian distribution fitted to the data.

where  $P_L$  and  $P_D$  are the probabilities of polymerization for L and D isomers, respectively.

Since crystal growth should be considered either as proceeding along planes or in three dimensions,  $\epsilon$  falls between:

$$2(P_L - P_D)/(P_L + P_D) \approx 2\sqrt{n} \epsilon_2 \tag{2}$$

and

$$2(P_L - P_D)/(P_L + P_D) \approx 2\sqrt[3]{n} \epsilon_3 \tag{3}$$

From the data,  $\epsilon_2 < 7 \times 10^{-13}$  and  $\epsilon_3 < 3 \times 10^{-10}$  was obtained. Keszthelyi (1977a) arrived at a similar figure by analyzing our first 63 crystallizations ( $\epsilon_2 < 1.6 \times 10^{-12}$  and  $\epsilon_3 < 5 \times 10^{-10}$ ).

These values are close to the theoretical value,  $\epsilon < 10^{-13}$  (Rein, 1974). Thiemann and coworkers (Thiemann, 1974), in their attempt to determine  $\epsilon$ , obtained  $\epsilon < 7 \times 10^{-6}$ , which is much larger and therefore calls for a different explanation.

## Discussion

## MECHANISM OF STEREOSELECTIVE INTERACTION

The irradiation-type experiments are generally rationalized on the basis of the Vester–Ulbricht process, i.e., stereoselective radiolysis of optically active solutes. The ratio of tartrates to water molecules in the saturated solution – before supersaturation takes place and crystallization begins – is about 1 : 10 which makes direct interaction between beta particles and dissolved tartrates a probable event in this system. The number of crystals deposited in each test tube by the end of the experiment was found to be about  $10^5$ – $10^6$ . As a comparison, one should bear in mind that  $10^8$ – $10^9$  beta particles are injected into the solutions each minute. Since crystal seeding takes days there is an overwhelming excess of betas (about  $10^{11}$ – $10^{12}$ ) with respect to the growing crystal nuclei ( $\approx 10^5$ ). Beta particles are not at all efficient seeding agents but a fraction of them can still contribute to nucleation. This is in line with the formation of more crystal seeds in the irradiated samples than in the controls.

The number of radiolyzed tartrates,  $N$  can be calculated from

$$N = I_0 \frac{E}{\delta} \tau (1 - e^{-T/\tau}) \quad (4)$$

where  $I_0$  is the initial radioactivity in the sample,  $E = 0.65$  MeV is the average energy of  $^{32}\text{P}$  beta electrons,  $\delta$  is the energy needed for a destructive interaction ( $\approx 30$  eV) (Keszthelyi, 1976b),  $T$  is the time of irradiation, and  $\tau$  is the half life of beta decay.

We found that only a very small fraction of the tartrates could be decomposed directly by this mechanism. These figures are given in Table III together with the maximum expectable optical activities. In the calculation the following assumptions were made: the radiolysis of water molecules is negligible, and the radiolysis of tartrates is 10% stereoselective.

Obviously, neither of the assumptions is rigorously correct; therefore the estimated optical activities are upper limits. Even in this case, the experimentally measured optical activities are about 10 times as large as the calculated ones. It is therefore concluded that

TABLE III  
Comparison of the estimated effect of stereoselective radiolysis with measured optical activities

Radioactivity (mCi/sample)	Doses (eV/g) $\times 10^{-18}$	Radiolyzed fraction (g/g tartrate) $\times 10^3$	Optical purity <sup>a</sup> (%)	Ratio of measured/ calculated <sup>b</sup> optical purities
5.0	144.4	1.52	0.24	16
1.6	46.2	0.49	0.18	36
0.1	2.9	0.03	0.04	13

<sup>a</sup>measured at 215 nm.

<sup>b</sup>estimated assuming 10% stereoselective radiolysis.

simple radiolysis of dissolved tartrates cannot be the main mechanism of the stereoselective effect. This is not very surprising if one considers that in this model each beta particle is assumed to give off its energy in 30 eV quanta, about  $2 \times 10^4$  decompositions per  $^{32}\text{P}$  beta particle. Only one of these hits can be stereoselective in which the spin of the beta electron is transferred. Consequently differential radiolysis cannot exceed  $10^{-4}$  in any experiment according to this mechanism. An increase in stereoselectivity will be achieved only if the circumstances are selected so that the effects of secondary processes are decreased (Zeldovich, 1974; Goldanskii, 1975).

Crystallization is an inherently heterogeneous, nonequilibrium process. Therefore the peculiarities of radiation chemistry become important here. Radiation chemical events differ from usual chemical reactions in the spatial distribution of reactive entities in the reaction mixture: the active species are produced along the track of an ionizing particle (spurs). Thus homogeneous reaction kinetics is hardly applicable to these systems (Draganic and Draganic, 1971). It is easy to see that spurs are larger and packed closer together at the end of the track. Mozumder and Magee (1966) determined the relative distances between spurs as well as their energy content. Three types of reactive units were found: (a) isolated spurs for which the deposited energy lies between 6 and 100 eV; (b) 'blobs' are the energy packages in the range of 100 eV to 500 eV; while (c) 'short tracks' represent a roughly cylindrical region where 500–5000 eV energy is transferred to the medium. Because of their different radical concentrations these units behave differently from a radiation-chemical point of view. For a  $^{32}\text{P}$  electron, out of the total energy (650 KeV) approximately 10% forms blobs, while in short tracks 23%, and in isolated spurs, 67% is released.

Part of the energy causes ionization; the remainder merely excites the molecules. Important information is the ratio of excitation to ionization. Calculations give values from 0.42 to 0.97 (Platzman, 1967; Santar and Bednár, 1969) i.e., roughly two-thirds of the energy is consumed to cause excitations.

Molecular aggregates, which will eventually grow to be crystal seeds, form with a much higher probability around an ionized solute molecule, because the charged molecule can orient and keep together the nearby nonionized ones. The highest concentration of ionized molecules will be in short tracks, thus that 7–8% (neglecting the effect of excitations) of the  $^{32}\text{P}$  beta energy plays the major role in the stereoselective crystal seed formation. Although occasionally fast secondary electrons are also produced in hard collisions, about 90% of secondary electrons have (Santar and Bednár, 1969)  $E < 500$  eV energies. Since there is an enormous supply of beta electrons in our system it can be assumed that the low energy secondary particles will not contribute significantly to crystal seeding because of their inability to form short tracks. Short tracks are also formed at the last stage of slow down of every primary beta electron. Therefore, a substantial part of short tracks are 'left-handed', i.e., due to spin polarization of its electron a stereoselective ionization of tartrates can take place. Thus a closer look at the fate of beta particles in aqueous solution revealed that the fraction of the beta energy that contains the spinpolarized primary beta particles ( $E < 5000$  eV) is crucial in crystal

seed formation, and much of the energy given off in the form of secondary beta particles is less efficient in nucleation.

Suppose the stereoselective cross section for differential seeding of tartrate crystallites is around 1% (Bonner *et al.*, 1974). If the crystals are more or less of the same size, an excess of 0.1% of one optical isomer should appear in the crystalline phase.

Of course, other seeding mechanisms (e.g., spontaneous nucleation, water radiolysis products, etc.) also operate. These effects increase the relative weight difference between control and irradiated crystallizations (about 20%) and decrease the optical purity. Both parameters are intimately linked to the beta irradiation of the crystallizing solution and from the measured data a stereoselectivity of 0.1% can be estimated. Furthermore, optically active tartrates have a solubility 5 to 6 times that of D,L-sodium-ammonium tartrate. This fact provides a pronounced amplification of both absolute and relative differences of the bias introduced in racemic conglomerate crystallization. Amplification permits either the relative effectiveness of short tracks or the 0.1% stereoselectivity to be smaller.

### Main Conclusions

Statistical analysis of a large number of independent experiments has established that both the weight increase and the stereoselective effect are significant and depend on the amount of radioactivity. Notably, the observed preferential crystallization effect seems to be large enough to overcome statistical fluctuations (Keszthelyi, 1976) or racemization (Fajszai and Czégé, 1977) during chemical evolution. For a more precise description these models of the Vester-Ulbricht process have to be extended to crystallization.

The stereoselective crystallization under the influence of  $\beta^-$  particles indicates a link between physical-crystal-biological asymmetries which offers the incorporation of many earlier findings into a complex picture (Morimoto *et al.*, 1973) (Figure 5). The results can be rationalized only by assuming a stereoselective interaction between spin-polarized beta particles and the dissolved optically active material. The efficiency of this process (*c* in Figure 5) is usually very small, but thanks to autocatalytic amplification it can be enhanced in crystallizations in the laboratory, or during chemical evolution as well (process *a* in Figure 5).

The observed asymmetric distribution of enantiomeric crystals in nature (Wald, 1957; Klabunovskii, 1960), becomes understandable, if the present results are extrapolated to an evolutionary scale. A sufficient amount of beta energy is known to have been present in the primordial environment (Keszthelyi, 1976a) which could have caused the stereoselective formation of asymmetric crystals.

The correct significance of stereoselective accumulation mechanisms involving asymmetric crystals (e.g., quartz) is recognized as soon as they need not be 'chance' processes. Thus, stereoselective crystallization induced by biased seeding (Harada, 1970) as well as stereoselective adsorption of enantiomers on asymmetric crystal surfaces (Morimoto *et al.*, 1973; Bonner *et al.*, 1974) represent particular steps through which physical asymmetry could have been transferred to the molecular level.

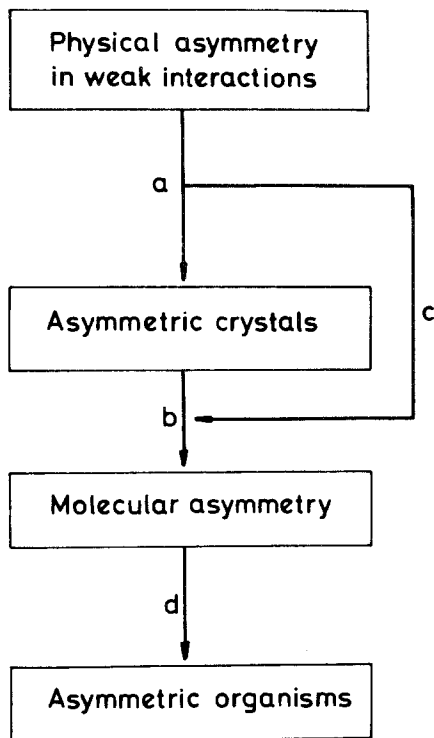


Fig. 5. The suggested relationship between asymmetries at different levels of organization.

It has also been suggested that clay and crystal surfaces could have catalyzed asymmetric prebiological chemical reactions, among others, polymerizations of amino acids (Bernal, 1967). These are again possible routes for appearance and amplification of asymmetry of parity violating forces via crystal asymmetry in biology (process  $a-b$  in Figure 5).

As indicated, some of the above results are at variance with the conclusions of Thiemann and co-workers (reviewed in Thiemann, 1974). No support was found in the present work in favor of any measurable energy content difference between optical isomers, starting either from symmetric or from non-symmetric conditions. The agreement within one or two orders of magnitude between theoretically estimated and experimentally found upper limits for the asymmetry effect,  $\epsilon$ , indicates that the direct asymmetric contribution is indeed as small as it has been predicted.

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