

β -Decay, Bremsstrahlen, and the Origin of Molecular Chirality¹

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Summary. A brief review is presented of the Vester–Ulbricht β -decay Bremsstrahlen hypothesis for the origin of optical activity, and of subsequent experiments designed to test it. Certain of our experiments along these lines, begun in 1974 and involving the irradiation of racemic and optically active amino acids in a 61.7 KCi ⁹⁰Sr–⁹⁰Y Bremsstrahlen source, have now been completed and are described. After 10.89 years of irradiation with a total Bremsstrahlen dose of 2.5×10^9 rads, crystalline DL-leucine, norleucine, and norvaline suffered 47.2, 33.6, and 27.4% radiolysis, respectively, but showed no evidence whatsoever of asymmetric degradation. D- and L-Leucine underwent about 48% radiolysis and showed 2.4–2.9% radoracemization. Other samples in solution were too severely degraded to analyze. Probable intrinsic reasons for the failure of the Vester–Ulbricht mechanism to afford asymmetric radiolysis in the present and related experiments involving β -decay Bremsstrahlen are enumerated.

Key words: Parity violation — β -Decay — Asymmetric radiolysis — Optical activity — Vester–Ulbricht hypothesis

Introduction

In the late 1950s, based on Lee and Yang's (1956) prediction of parity violation during β -decay, F. Vester (Vester 1957; Vester et al. 1959) and T. L.

V. Ulbricht (Ulbricht 1959; Ulbricht and Vester 1962) proposed the following hypothesis for the origin of the singular chirality found in the contemporary biosphere: radionuclide —(β -decay)→ longitudinally polarized electrons —(deceleration)→ circularly polarized Bremsstrahlen —(asymmetric photochemical interaction with organic matter)→ chiral organic molecules. Each step in the Vester–Ulbricht (V-U) mechanism has in principle received experimental confirmation. Thus Wu and coworkers (1957) demonstrated that β -decay electrons were indeed longitudinally polarized with a predominant antiparallel (“left-handed”) spin. Goldhaber et al. (1957) showed that the high energy x-ray Bremsstrahlen produced on deceleration of such chiral electrons were similarly chiral, i.e., were “left-handed” circularly polarized photons. The final asymmetric photochemical link in the V-U mechanism was originally demonstrated (by longer-wavelength analogy) in the classic circularly polarized ultraviolet photolyses of Kuhn and Braun (1929) and Kuhn and Knopf (1930a, b), and in the subsequent ultraviolet photolytic or photosynthetic asymmetric reactions of Kagan and coworkers (1971), Bernstein et al. (1973), and others. No asymmetric effects have been demonstrated, however, with circularly polarized photons of shorter than ultraviolet wavelengths.

Vester et al. (1959) and Ulbricht and Vester (1962) attempted to test their hypothesis experimentally by conducting, in the presence of a wide variety of β -emitters, a number of organic reactions that could yield chiral products. The products of 36 such experiments were examined for optical activity using precision polarimeters, but all were found to be optically inactive within experimental error. Shortly thereafter Gol'danskii and Khrapov (1963) reported that no optical activity was induced on irradiating ten racemic samples (including eight amino acids)

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with β -rays from a ^{104}Rh source, nor were differences noted in the effects of such radiation on optically pure antipodes. In 1968 Garay reported the first positive result in such experiments, claiming that D-tyrosine in alkaline aqueous ethanol was more decomposed than was L-tyrosine after an 18-month exposure to both the β -rays and Bremsstrahlen from 0.36 mCi of codissolved $^{90}\text{SrCl}_2$. Garay's criteria for asymmetric degradation in these experiments were differences in the ultraviolet absorption spectra of the D- and L-tyrosine samples after 18-month exposure, the 250- and 300-nm absorption maxima being noticeably more degraded for the D-tyrosine sample.

Because of the importance of Garay's observations for the potential validity of the V-U mechanism, we felt it desirable to modify and extend this type of experiment to include other amino acids, and other and more powerful β -ray (or Bremsstrahlen) sources, and in particular to employ crystalline target substrates, thereby obviating the potentially obscuring effects of aqueous solution radiochemistry involving symmetrical free radicals. In all such experiments over the past decade, our general procedure has been to subject crystalline and dissolved DL-amino acids to partial (50–70%) radiolysis in the β -ray or Bremsstrahlen source being employed, and then to examine the residual undegraded amino acid in the product for its enantiomeric composition using analytical gas chromatography (GC). The advantages of using GC as a criterion for optical activity and for establishing percentage radiolysis have been fully described (Bonner 1973; Bonner et al. 1974; Bonner and Blair 1979; Blair and Bonner 1980a).

Our initial experiments (Bonner 1974; Bonner and Flores 1975) used a 61,700-Ci ^{90}Sr – ^{90}Y Bremsstrahlen source located at Oak Ridge National Laboratory. Three containers, each holding 21 identical vials containing separate amino acid samples, were immersed in this source, the containers were retrieved after increasing radiation exposures, and the samples were examined for optical activity. The first container was retrieved after 124 days (dose: 1×10^8 rads) and the second after 1.35 years (dose: 4.1×10^8 rads). The amino acids examined from each container proved to be significantly radiolyzed, but the enantiomeric compositions of all samples proved to be 50:50 (D/L) within experimental error. Retrieval of the third container after 10.9 years (dose: 2.5×10^9 rads) and examination of certain of its amino acid samples is the subject of the present article.

Since our initial experiments, we have submitted other amino acids to irradiations with electrons and Bremsstrahlen from two additional natural β -emitting radionuclides, ^{14}C and ^{32}P . In 1978 (Bonner et al. 1978a) we examined six racemic and seven optically active ^{14}C -labeled crystalline amino acids that had been prepared 17–25 years earlier (Bernstein et al. 1972) and had received radiation doses of 5 – 11×10^7 rads. Despite 26–68% gross degradation, the racemic samples showed no asymmetric radiolysis [i.e., showed 50:50 (D/L) enantiomeric composition]. Encouraged by a report by Darge et al. (1976) that a 19% optical enrichment accompanied the 33% gross radiolysis of DL-tryptophan with 5 mCi [^{32}P]phosphate in frozen aqueous solution (2 ml, -25°C , 12 weeks), we reinvestigated the possible asymmetric radiolysis of both DL-tryptophan and DL-leucine with ^{32}P β -rays and Bremsstrahlen (Bonner et al. 1979a; Blair and Bonner 1980b). However, both under Darge's exact conditions and under lower-temperature ($\sim 196^\circ\text{C}$) anhydrous conditions, and using GC rather than polarimetry as the criterion for optical activity, we found no asymmetric degradation whatsoever in any of the ^{32}P -radiolyses that we conducted.

Failing to induce asymmetric decomposition after 1.35 years using the above 61.7 kCi ^{90}Sr – ^{90}Y Bremsstrahlen source, we undertook to attempt asymmetric radiolyses using longitudinally polarized electrons directly, rather than their Bremsstrahlen. In these experiments DL-leucine was irradiated in a 120-keV linear accelerator capable of producing either natural, antiparallel-spin (AP), "left-handed" electrons or parallel-spin (P) electrons of 13–32% net polarization. After 53–76% gross radiolysis, AP electrons yielded residual leucine having a 0.60–1.42% enantiomeric excess (EE) of the L-enantiomer, while P electrons afforded a residue having a 0.74–1.14% EE of the D-isomer (Bonner et al. 1975, 1976/77). Theoretical aspects of these experiments have been discussed more recently (Keszthelyi 1976; Walker 1976; Bonner et al. 1976, 1978b; Noyes et al. 1977). For reasons not yet understood, a careful attempt to duplicate the above asymmetric radiolyses using a different source of AP and P polarized electrons has been unsuccessful (Bonner et al. 1979b; Hodge et al. 1979). As none of the positive reports for asymmetric β -radiolysis of amino acids has been confirmed, we have since attempted analogous experiments with yet another "artificial" chiral particle, namely, P and AP longitudinally polarized protons produced in a cyclotron. Using P or AP protons of 0–10 MeV and radiation doses of 1 – 9×10^8 rads, and obtaining gross degradations of 7–50%, however, we again found no asymmetric effects in a variety of such irradiations involving crystalline DL-leucine (Lemmon et al. 1981).

In our ^{14}C -labeled amino acid studies (Bonner et al. 1978a) we noted a phenomenon having a potentially deleterious effect on the V-U mechanism, namely, that the radiolysis of optically active amino acids was accompanied by small amounts of race-

mization of the undecomposed amino acid residues. Using up to 10^9 -rad doses of γ -radiation from a 3000-Ci ^{60}Co source, we have since shown that such "radiatoracemization" is a general phenomenon for both crystalline amino acids and their dissolved sodium (but not HCl) salts (Bonner and Lemmon 1978a, b). Radoracemization was also noted previously in the ^{90}Sr - ^{90}Y radiolyses of D- and L-leucine (Bonner 1974), and subsequently in direct irradiations of these isomers with 0–11 MeV protons (Bonner et al. 1982). Some geochemical and cosmochemical implications of radoracemization have recently been pointed out (Bonner et al. 1979c, d, e), and its potential jeopardizing effect on the efficacy of the V-U mechanism for the origin of optical activity has been stressed (Bonner and Lemmon 1978b).

Experimental

β -Ray Bremsstrahlen Source. The source employed, located at the Oak Ridge National Laboratory (ORNL), consisted originally (January 1970) of a total of 61.7 kCi ^{90}Sr - ^{90}Y oxide in four sealed, round, stainless steel cans 6.4 cm (diameter) \times 23 cm (high) situated at the corners of an 18.4-cm² area and immersed under 3 m of water, as previously described (Bonner 1974). Samples indicated below were placed in the center of the source in mid-March 1971, when the remaining radioactivity was 59.9 kCi and the calibrated dose rate (Isotopes Division, ORNL) was 3.55×10^4 rads/h. In mid-January 1976 all but 38.0 kCi of the ^{90}Sr - ^{90}Y was removed from the source, providing a new dose rate of 2.25×10^4 rads/h. The irradiation was terminated on February 2, 1982, after 10.89 years and a total Bremsstrahlen dose of approximately 2.5×10^9 rads.

Samples Irradiated. Three 5.1 \times 8.3 cm (diameter) sealed aluminum cans, each holding twenty-one 5-ml vials containing samples, were placed in a lead-weighted stainless steel cage, which was then immersed in the center of the Bremsstrahlen source; the three sets of 21 samples were identical. The samples consisted primarily of crystalline D-, L-, and DL-amino acids, as well as their neutral, acidic, and alkaline aqueous solutions. Retrieval of two of the containers of samples after 124 days (1.0×10^8 rads) and 494 days (4.1×10^8 rads), respectively, and examination of certain of the samples in each for optical activity (either by optical rotation or by GC) and for percentage radiolysis (by GC) has been fully described before (Bonner 1974). The following describes our examination of certain of the samples remaining in the third container, which was removed from the Bremsstrahlen source after 10.89 years (2.5×10^9 rads).

Analysis of Samples. Of the 21 samples originally placed in the Bremsstrahlen source, the only ones that were not obviously excessively decomposed after 10.89 years were the crystalline amino acids. Accordingly, these received our main attention. A weighed sample of each irradiated amino acid (ca. 9.0 mg) was dissolved in a few milliliters of 4.5 M HCl/2-propanol reagent, and the solution was diluted to 10.00 ml with additional reagent. Then 5.00 ml of the diluted solution was added to the appropriate "enantiomeric marker" (ca. 5.0 mg) for the amino acid in question to determine percentage radiolysis by the enantiomeric marker method (Bonner 1974; Blair and Bonner 1980a). The solutions were refluxed for 3 h, then stripped of solvents under vacuum. The residues were dissolved in 2 ml dichloromethane mixed with 1 ml trifluoroacetic anhydride. After 0.5 h the solvents were evaporated and the 2-propyl N-trifluoroacetyl-amino acid ester residues were dissolved in ca. 2 ml dichloromethane for GC analyses of their enantiomeric compositions. GC analyses were conducted using 150 foot \times 0.02 in. inner diameter stainless steel capillary columns coated with either N-docosanoyl-D-valine tert-butylamide or the corresponding L-valine derivative (Bonner and Blair 1979) and installed in a Hewlett-Packard model 5700A gas chromatograph coupled to a Hewlett-Packard model 3380A digital integrator. All analyses of irradiated samples were interspersed back to back with analyses of nonirradiated control samples prepared from the same original amino acid source (Bonner 1974). Details of these analyses are shown in Table 1.

Solutions of D- or L-leucine (65 mg) in 4 ml 9:1 water-ethanol, irradiated as above, were evaporated to dryness and the residues were derivatized and analyzed by GC. No leucine could be detected in the residues. A sample of D-leucine in the 9:1 water-ethanol solvent that had been irradiated for 1.35 years (4.1×10^8 rads) (Bonner 1974) was then analyzed and found to be 98% decomposed as well as significantly racemized (%D, 86.64; %L, 13.36). Other dissolved samples (Bonner 1974) were not analyzed due to their excessive radiolysis.

Discussion

In our previous analyses of DL-leucine irradiated with ^{90}Sr - ^{90}Y Bremsstrahlen for 1.35 years (Bonner 1974) the average enantiomeric composition was %D, 50.42, and %L, 49.58 ± 0.29 , while the control showed %D, 50.18, and %L, 49.82 ± 0.31 , a difference of 0.24. These are comparable within experimental error to the present analyses for irradiated DL-leucine and its control in Table 1. Clearly there was no evidence for asymmetric radiolysis of DL-

Table 1. ^{90}Sr - ^{90}Y Bremsstrahlen radiolyses of several crystalline amino acids^a

Amino acid	Irradiated sample					Control sample				Differ- ence ^b
	No. of anal.	%D	%L	SD ($\pm\%$)	% decomp.	No. of anal.	%D	%L	SD ($\pm\%$)	
DL-Leucine	8	50.37	49.63	0.34	47.2	11	50.28	49.72	0.22	0.09
DL-Norleucine	5	50.14	49.86	0.31	33.6	5	50.23	49.77	0.24	-0.09
DL-Norvaline	9	49.88	50.12	0.15	27.4	9	50.12	49.88	0.19	0.24
D-Leucine	5	98.48	1.52	0.09	49.5	5	99.70	0.30	0.07	-1.22
L-Leucine	5	1.64	98.36	0.28	47.9	5	0.16	99.84	0.15	-1.48

^a Total dose: ca. 2.5×10^9 rads^b % of most prevalent isomer (irradiated) - % of corresponding isomer (control)**Table 2.** Radiolysis and radoracemization of leucine with varying radiation sources and doses^a

No.	Radiation	Dose (rads $\times 10^8$)	% Radiolysis (A)	% Radorace- mization (B)	(A per rad) $\times 10^8$	(B per rad) $\times 10^9$
1	^{60}Co γ -rays	8.1 ^b	68.0	5.3	8.4	6.5
2	^{60}Co γ -rays	10.2 ^b	94.7	10.6	9.3	10.0
3	^{90}Sr - ^{90}Y Bremsstrahlen	4.1 ^c	14.5	1.2	3.5	2.9
4	^{90}Sr - ^{90}Y Bremsstrahlen	26 ^d	48.7	2.7	1.9	1.0
5	Cyclotron protons	5 ^e	47.1	1.5	9.4	3.0

^a Average values for D- and L-leucine in the references cited^b Bonner and Lemmon (1978a, b)^c Bonner (1974)^d Present work^e Bonner et al. (1982)

leucine after 1.35 years, nor is there now, after 10.89 years. The same conclusion is evident from the data in Table 1 for DL-norleucine and DL-norvaline, in which the enantiomeric compositions of the irradiated and control samples are again indistinguishable within experimental error. It is interesting that the DL-norvaline and DL-norleucine in Table 1 are only 27-33% decomposed, whereas the D-, L-, and DL-leucine samples are 47-49% destroyed with the 2.5×10^9 -rad Bremsstrahlen dose employed. This accords with the relative radiation stabilities of these amino acids as seen in ^{60}Co and ^{137}Cs γ -radiolyses (Tolbert et al., 1962, unpublished manuscript; Bonner and Lemmon 1978a, b). It is also noteworthy that the present ^{90}Sr - ^{90}Y Bremsstrahlen radiolyses are less efficient than were ^{60}Co γ -radiolyses. Thus 8.1×10^8 rads of γ -radiation gave 68% radiolysis of D- and L-leucine (Bonner and Lemmon 1978b) as compared with 48-49% radiolysis with 2.5×10^9 rads of Bremsstrahlen in the present case.

Comparison of the irradiated and control sample data for D- and L-leucine in Table 1 indicates that the D-leucine underwent about 2.4% radoracemization during its 49.5% radiolysis, while the L-leucine suffered 2.9% racemization on 47.9% radiolysis (average: 2.7% radoracemization for 2.5×10^9 rads exposure). It is interesting to compare these values with the efficacies of radiolysis and radoracemi-

zation observed with several of the radiation sources we have employed in the past. In Table 2 are listed the average values for the extents of radiolysis and radoracemization for crystalline D- and L-leucine for varying doses of ionizing radiation from several sources, as well as the percentages of degradation and racemization per unit of dose. We see that ^{60}Co γ -radiation is considerably more effective than ^{90}Sr - ^{90}Y Bremsstrahlen in causing both radiolysis and radoracemization, whereas cyclotron protons give more efficient radiolysis than do the Bremsstrahlen, but similar radoracemization. The lower efficiency of the ^{90}Sr - ^{90}Y Bremsstrahlen may be attributable in part to uncontrolled dose variations with the ^{90}Sr - ^{90}Y source employed. In any case, the small amounts of racemization attending the significant radiolyses of leucine with ^{60}Co γ -rays, ^{90}Sr - ^{90}Y Bremsstrahlen, or cyclotron protons make it appear highly unlikely that radoracemization could have been an important factor in our failure to detect asymmetric radiolysis of crystalline amino acids with either longitudinally polarized protons (Lemmon et al. 1981) or with ^{90}Sr - ^{90}Y Bremsstrahlen.

Since radoracemization appears to be at best only a secondary factor, it seems probable that the primary reason for the failure of the V-U mechanism to provide asymmetric radiolysis in the present experiments has to do with intrinsic properties of the

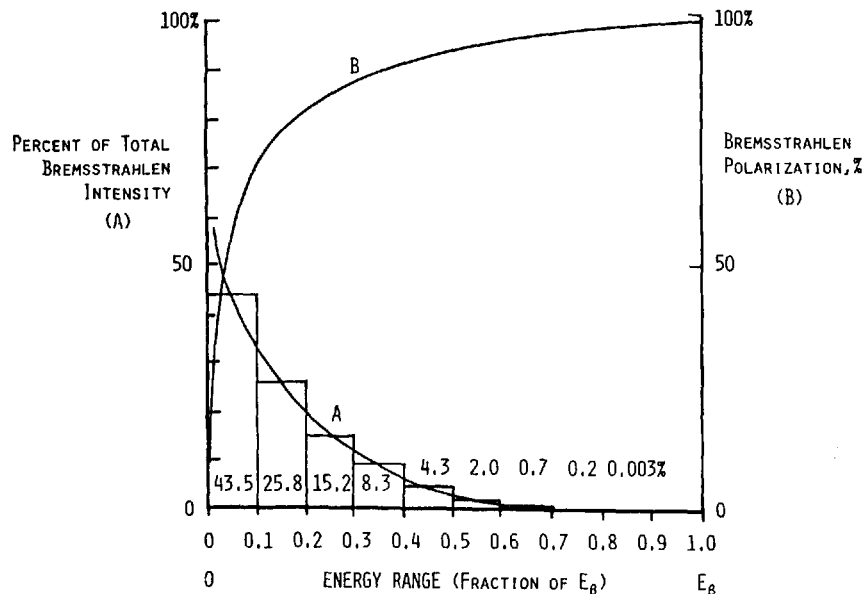


Fig. 1. Bremsstrahlen intensity and circular polarization as a function of energy. E_β , maximum energy of β -decay electrons. [Adapted from Wyard (1955) and McVoy (1957)]

Bremsstrahlen themselves. As seen in Fig. 1, Bremsstrahlen photons embrace a spectrum of energies from zero up to the maximum energy of the β -decay electrons, with the vast majority of the Bremsstrahlen intensity lying in the low-energy region of the spectrum (Wyard 1955). Figure 1 also shows that the circular polarization of the Bremsstrahlen produced by longitudinally polarized electrons ranges from zero at the low-energy end to almost 100% at the high-energy end of the Bremsstrahlen spectrum (McVoy 1957; Fronsdal and Überall 1958; Schopper and Galster 1958). Thus the vastly predominant low-energy Bremsstrahlen photons, with energies in a range most capable of engendering simple photochemical processes, are too insufficiently circularly polarized to produce asymmetric photochemical effects of observable magnitude. At the same time, the more completely polarized high-energy Bremsstrahlen photons are both too few and in the wrong energy domain to produce asymmetric photochemical changes. Both Keszthelyi (1976) and Walker (1976) have argued on theoretical grounds that the asymmetric radiolyses reported with longitudinally polarized linear accelerator electrons (Bonner et al. 1976/77) could not have been caused by the accompanying Bremsstrahlen, and Walker (1976) has emphasized that low-energy, photochemically efficient Bremsstrahlen photons would be stopped by the container material in experiments such as the present ones. Furthermore, Keszthelyi and Vincze (1975) found no differences within experimental error for the absorption by several D- and L-amino acids of right- and left-circularly polarized γ -radiation in the low-energy Bremsstrahlen range (14.4 keV), which would

preclude asymmetric decomposition by photons of this energy. Finally, in Bremsstrahlen radiolyses in aqueous solution, extensive radoracemization (as seen above) and symmetrical reactions induced by free radicals from solvent radiolysis would effectively prevent any observable asymmetric phenomena involving racemic or prochiral substrates. In view of the consistent experimental failures and theoretical obstacles cited above, it appears likely that the V-U hypothesis involving β -ray Bremsstrahlen as the agent responsible for the primordial origin of optical activity is experimentally unverifiable and probably operationally invalid [cf. also Walker (1976)].

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