the presumed compensatory effect in some of the D. virilis hosts, the nature of this effect is being further investigated. The species difference between D. melanogaster and D. virilis in the appearance of the effect is especially provocative (7).

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# **Radiation-Induced Reaction of Carbon Dioxide with Ethylene**

Abstract. The gamma irradiation of mixtures of carbon dioxide with ethylene vielded long-chain carboxylic acids. The G(-ethylene) values varied with reaction temperature from essentially zero at -78°C to 175 at 100°C. Under the same conditions, the substitution of free radical agents for radiation gave polyethylene.

A new route to long-chain carboxylic acids has been found in the radiationinduced reaction of ethylene with carbon dioxide. Sargent (1) has reported the formation of a "carbon dioxidemodified polyethylene" by heating ethylene in the presence of carbon dioxide and a free radical-producing catalyst. The comparatively strenuous reaction conditions (benzoyl peroxide, 9000 to 15,000 lb/in.2 (gage), at 50° to 300°C) are reminiscent of those employed in the preparation of conventional or low density polyethylene. It is apparent from this reference that very little carbon dioxide was incorporated into the product. In the experiments discussed in this report, carbon dioxide-ethylene products were formed under gamma radiation at comparatively low pressures and temperatures (see Table 1). The importance of radiation was shown in two experiments under these conditions of temperature and pressure in which free radical initiators, di-tert-butyl peroxide and 2,2'-

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azobis(2-methylpropionitrile), were substituted for irradiation. The products were observed, by infrared spectra on each product and by carbon, hydrogen analyses on the product of the peroxideinduced reaction, to be only polyethylene.

Data listed in Table 1 clearly indicate that radiation yield, measured as G for the disappearance of ethylene (the number of molecules of ethylene consumed per 100 ev of energy absorbed), rises with rising temperature but that carbon dioxide incorporation, measured by carbon, hydrogen analyses, declines with rising temperature.

The products are white, waxy solids at temperatures between 50° and 90°C. Their infrared spectra are consistent with a linear carbon chain possessing points of unsaturation, crystalline polyethylene structure, and carboxyl groups. The failure of the products to react with 2,4-dinitrophenyl hydrazine in amounts detectable by infrared analysis (through the strong absorptions of the nitro group) indicates the absence of aldehydes and ketones. The infrared spectrum of the sodium salt includes absorption bands characteristic of RCOONa. This salt was prepared by shaking a benzene solution of the carbon dioxideethylene product with 10-percent aqueous sodium hydroxide. The resulting emulsion did not break in 72 hours. For the three products on which both number average molecular weight and carbon, hydrogen analyses are available, the carbon, hydrogen data closely approach agreement with those calculated for a simple carboxylic acid of the experimentally determined molecular weight. These acids may be formed by combination of a growing polyethylene chain with a carbon dioxide molecule to yield a resonance stabilized radical that is not sufficiently energetic to react by further chain growth.

$$\begin{array}{c} 0\\ --CH_{2}CH_{2} \cdot + CO_{2} \rightarrow --CH_{2}CH_{2}C \\ 0 \cdot \\ 0 \cdot \\ \leftarrow \rightarrow --CH_{2}CH_{2}C \\ 0 \end{array}$$

This would appear to preclude polyester formation by continued reaction with ethylene. Since the infrared spectra of high molecular weight acids and esters are similar, a sample was examined by nuclear magnetic resonance (NMR)

spectrometry. Had the -C-O- content indicated by carbon, hydrogen data exceeded the acidic hydrogen content determined by NMR spectrometry, one might surmise that the excess is present as polyester linkages. However, the acidic hydrogen content was too low for the NMR spectrogram to be significant. A conventional saponification equivalent determination failed to detect hydrolyzable linkages. In view of the

Table 1. Data on the formation of carbon dioxide-ethylene products with gamma irradiation and with free radical initiators. The appearance of the products is shown in the last column by the abbreviations: T.L., tan liquid; W.P., white paste; and T.W.P., thin white paste.

| Reactants   | Charge<br>(g)        | Temp.<br>(°C) | Yield<br>(g) | G<br>(-C <sub>2</sub> H <sub>4</sub> ) | Mol.<br>wt.* | Analyses |      |       |      |         |
|---|----------------------|---------------|--------------|--|--------------|----------|------|-------|------|---------|
|   |                      |               |              |  |              | Calcd.†  |      | Found |      | Product |
|   |                      |               |              |  |              | C        | н    | C     | Н    |         |
| CO <sub>2</sub><br>C <sub>2</sub> H <sub>4</sub>                                    | · 29.9<br>140.1      | - 78          | None         |  |              |          |      |       |      |         |
| $CO_2$<br>$C_2H_4$  | 24.2<br>140.1        | -40           | 0.1          | 2.3                                    |              |          |      |       |      | T.L.    |
| $\begin{array}{c} CO_2 \\ C_2H_4 \end{array}$                                       | 20.0<br>139.8        | 29            | 1.7          | 39.4                                   | 568          | 81.0     | 13.5 | 80.1  | 12.0 | W.P.    |
| $\begin{array}{c} \mathrm{CO}_2 \\ \mathrm{C}_2\mathrm{H}_4 \end{array}$            | 21.2<br>146.2        | 65            | 1.9          | 42.0                                   | 574          | 81.3     | 13.5 | 81.6  | 13.4 | W.P.    |
| $\begin{array}{c} CO_2 \\ C_2H_4 \end{array}$                                       | 19.8<br>142.4        | 100           | 7.7          | 175.0                                  | 670          | 81.6     | 13.6 | 83.6  | 13.9 | T.W.P.  |
| $CO_2$<br>$C_2H_4$  | 21.9<br>140.8        | 204           | 13.3         | 310.0                                  | 660          |          |      | 85.0  | 14.5 | T.L.    |
| CO <sub>2</sub><br>C <sub>2</sub> H <sub>4</sub><br>( <i>t</i> -BuO) <sub>2</sub> ‡ | 20.3<br>140.3<br>1.0 | 125           | 4.51         |  | 1150         | -        |      | 85.6  | 14.5 | W.P.    |
| CO <sub>2</sub><br>C <sub>2</sub> H <sub>4</sub><br>ABMP¶                           | 20.0<br>139.5<br>1.0 | 85            | 0.42§        |  |              |          |      | 11    |      | W.P.    |

\* Determined by benzene boiling point elevation. \*Calculated for a saturated monocarboxylic acid of experi-Betalinder noise a saturated noise a saturated noise a saturated noise a boxyle act of experi-mentally determined molecular weight.  $\ddagger$  Di-*iert*-butyl peroxide, Shell Chemical Corp. \$ Corrected to ex-clude weight of initiator residue.  $\parallel$  Product contaminated by initiator residue.  $\P$  2,2'-Azobis(2-methylpropionitrile), Eastman (white label).

coincidence of carbon, hydrogen and molecular weight data, which are consistent with the concept of the product type being a simple, long-chain carboxylic acid, it seems unlikely that much, if any, polyester structure is present.

The reactants were charged to reaction vessels (American Instrument Co. Micro Series) having an approximate volume of 300 ml. Carbon dioxide was added in the solid phase and ethylene (99.5-percent purity) was quickly pressured into the bombs while still cold. Calculated initial pressures ranged from 1000 to 7500 lb/in.<sup>2</sup> (gage), depending upon the temperatures employed. Gamma irradiations were made (see Table 1) to an approximate total dose of  $1 \times 10^7$  r with a 6-Mev linear electron accelerator as an x-ray source (2). In experiments in which free radical initiators were used in lieu of radiation, the reaction was carried out at the decomposition temperature of the initiator. C. E. Stoops

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### **References and Notes**

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- The gamma irradiation was conducted at either the MTR Gamma Facility, Materials Testing Reactor, Idaho Falls, Idaho, or at the Radia-tion Laboratory, Phillips Research Center, Bartlesville, Oklahoma.

15 May 1961

## **Tonus of Extrinsic Laryngeal Muscles during Sleep and Dreaming**

Abstract. The tonus of extrinsic laryngeal muscles was studied in sleeping humans by means of electromyograms. A striking decrease in the muscle tonus was observed at the onset of each phase of electroencephalographic light sleep, rapid eye movements, and dreaming.

Previous electromyographic (EMG) studies of muscle tonus during sleep in humans have found the resting potential to decrease when the subjects fall asleep, but no relationship was found between resting potential and the depth of sleep determined from the electroencephalograph (EEG) (1).

The aim of the present study was to investigate the high resting tonus of the laryngeal muscles during continuous sleep. Brain waves, eye movements, and electromyograms were recorded, by essentially the same procedure as that of Dement and Kleitman (2), in nine sub-

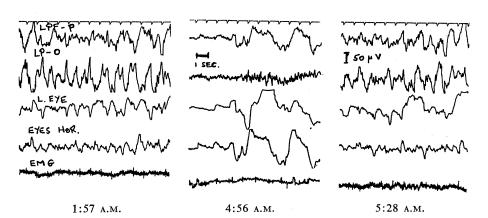


Fig. 1. Variation in laryngeal resting potential with depth of sleep. The EMG activity recorded during ocular quiescent deep sleep phases at 1:57 A.M. and 5:28 A.M. is markedly greater than that recorded at 4:56 A.M. during the intermediate dream period of light EEG sleep, with rapid eye movements clearly seen in the eye channels.

jects for a total of 17 nights of continuous sleep.

The electroencephalogram was recorded by means of an anteroposterior chain of electrodes affixed to the scalp with collodion. Eye movements in the horizontal plane were recorded by the electro-oculogram, by a central forehead electrode and a lateral pair made up of an electrode at each outer canthus, attached to the surface of the skin by means of sticking plaster.

Electrical activity in the laryngeal musculature was monitored by surface electrodes attached to the skin with sticking plaster over the supra- and infrahvoid muscles.

When the subjects fell asleep there was invariably a continual, discrete decrease in the EMG activity, until the appearance of sleep spindles and Kcomplexes. The EMG activity then tended to remain constant, apart from momentary increases associated with gross body movements, swallowing, or localized movements. These could be identified by means of a sensitive microphone at the head of the bed and by muscle artifact in the EEG and eye leads.

The depth of sleep, as indicated by the EEG, showed the typical cyclical pattern (3), with rapid eye movements occurring during the light phases of "A" and "B" stage sleep.

A rapid decrease in EMG activity was invariably observed, with the disappearance of sleep spindles, on the upswing of a sleep cycle from the "D" and "E" stages of deep sleep, through the "C" stage to the "A" and "B" stages, accompanied by rapid eye movements normally associated with dreaming. An example of this variation in EMG activity is shown in Fig. 1.

Jouvet et al. (4) have noted a similar

variation of EMG activity in the nuchal muscles of cats during sleep. After a "slow" stage of sleep characterized by spindles and slow waves, there was a "rapid" sleep phase: the "paradoxical phase" which was characterized by a rapid, low-voltage, nonspindling electroencephalogram similar to that of wakefulness, accompanied by rapid eye movements and total disappearance of EMG activity. This phase is identified by Jouvet (in 5, p. 204) with the "A" and "B" sleep stages, rapid eye movements, and dreaming in humans. This view has been contested by Schwartz and Fischgold (in 5, pp. 225, 232), who maintain that there is an increase in oropharyngeal muscle tone with eye movements, on the grounds that snoring occurs extremely rarely during rapid eye movement periods, snoring being attributed to the loss of muscle tone in the soft palate.

It therefore appears from the present study either that the "A" and "B" sleep stages in humans are equivalent to Jouvet's "paradoxical phase" of "rhombencephalic sleep," or that the observed variation of muscle tonus during sleep is peculiar to the laryngeal muscles.

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