

to parallel the presumed change of farming economy between stages A and B. The surplus labor required for the construction of the great megalithic tombs could have been connected with a change to more productive farming methods such as might have happened in stage B. The oldest radiocarbon dates so far obtained from an Irish megalith are 2925 ± 150 B.C. (sample UB-318) and 2845 ± 185 B.C. (sample UB-319) for charcoal from soil beneath one of the satellite graves at Knowth, County Meath (15). Clearly much more dating, particularly of megaliths, will have to be done before any such connection can be established, and it is to be expected that different economic patterns will be demonstrated in different regions.

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Preparation of High-Crystallinity Polyethylene at Low Pressures

Abstract. *The preparation of highly crystalline polyethylene at low pressures is reported. With careful control of the substrate, the melting and crystallization schedule, and the physical state of the specimen (film thickness), it is possible to prepare polyethylene having a density in excess of 0.999 gram per cubic centimeter, a melting temperature of approximately 140°C, and a heat of fusion in excess of 70 calories per gram. The approach appears to be general and should be applicable to a wide variety of polymers.*

We report here the preparation at reduced pressures of polyethylene having a final melting temperature in excess of 139°C, a density in excess of 0.999 g/cm³, and a heat of fusion, in the melting region, in excess of 70 cal/g. Although there is an extensive body of literature (1, 2) on the preparation of high-pressure crystallized polyethylene possessing both a high melting temperature ($\sim 140^\circ\text{C}$) and a high heat of fusion ($\Delta H_f \cong 70$ cal/g), few data exist to demonstrate the possibility of preparing such a high-crystallinity polyethylene at normal or reduced pressures (3).

We have paid particular attention to the method of sample preparation, especially the substrate (nucleating surface), the detailed thermal history, and the film thickness of the polyethylene. It has already been demonstrated (4) that the substrate has a profound effect on the interfacial properties of the poly-

mer, as evidenced by changes in morphology (5), mechanical properties (6), and wettability (5, 7). In order to observe these effects it is imperative that the polymer thermodynamically spread (contact angle equals zero) on the substrate used to generate the polymer and that the substrate be removed from the polymer by dissolution rather than by mechanical means. When extremely thin sections are examined calorimetrically, the substrate may be left in contact with the polymer.

Two linear polyethylenes (unfractionated Marlex 6050 and a low-molecular-weight narrow fraction of Marlex 6001 with an average molecular weight of 10,000) were used in this study. A gold foil (0.5 mil, 99.999 percent pure) was used as the nucleating surface.

There is dual purpose in selecting gold as the nucleating substrate for polyethylene. Absence of an oxide

coating not only aids in precluding oxidation of the polymer but also yields a polyethylene having the highest interfacial density (5, 7).

Polyethylene films between 0.2 and 1.5 mils thick were prepared by molding composites consisting of gold-polyethylene-gold at 1000 pounds per square inch (68 atm) and 170° to 200°C for 30 minutes. The composites were cooled rapidly by circulating water through the press platens. The rate of cooling was in excess of 20°C/min. Thin polyethylene sections (0.10 to 0.15 mil) were prepared by casting from a solvent a hot xylene solution of polyethylene (1.8 percent) on gold. The major portion of the solvent was evaporated above the melting point of the polyethylene in a nitrogen atmosphere. The gold-coated polymer was stored under vacuum for a minimum of 1 week to remove residual xylene. The final composite was formed by placing an additional piece of gold film on the polymer-coated foil and molding at 170°C for 30 minutes after which the composite was cooled according to the cooling schedule given above. Disks consisting of gold-polyethylene-gold composite (0.634 cm in diameter) were punched out with a special die for use in the differential scanning calorimeter (Perkin-Elmer model DSC-1B). The samples were heated and cooled in a programmed fashion under a reduced nitrogen pressure. Specimens prepared at atmospheric nitrogen pressure showed slight signs of oxidation, as evidenced from an infrared examination. The use of a reduced pressure of nitrogen (0.25 mm-Hg) is sufficient to preclude the oxidation noted in our earlier experiments. The results reported here are for polyethylene prepared at reduced pressures.

The differential scanning calorimeter was modified with a repeat cycle timer (G. C. Wilson and Co.) to extend the rate of heating or cooling of the specimens from 0.625°C/min to 0.001°C/min. The density gradient column contained a mixture of diethylene glycol and isopropyl alcohol (1:1), which was maintained at 23°C and 50 percent relative humidity.

The thickness of the samples was measured with an electronic micrometer (J. W. Dial & Co.) and also calculated from the dimensions, the weight of the sample, and the measured density. When the density was not measured, an arbitrary value of 0.975 g/cm³ was used. The film thicknesses

Table 1. Change in the heat of fusion of gold-nucleated polyethylene, Marlex 6050, as a function of the rate of cooling from the melt (142°C).

| Film thickness (mil) | Weight of sample (mg) | Rate of cooling (°C/min) | Heat of fusion, $H\Delta_f$ (cal/g) | Density, ρ , at 23°C (g/cm ³) | Melting temperature (final) (°C) | % Crystallinity (θ) | | |
|----------------------|-----------------------|--------------------------|-------------------------------------|------------------------------------------------|----------------------------------|------------------------------|--------------------|---------------------|
| | | | | | | θ_1^* | θ_2^\dagger | θ_3^\ddagger |
| 1.42 | 1.109 | 0.004 | 62.2 | | 138.3 | 88.9 | | |
| 1.38 | 1.077 | .004 | | 0.9895 | | | 86.6 | 93.7 |
| 1.43 | 1.121 | .008 | 62.0 | | 138.3 | 88.6 | | |
| 1.32 | 1.029 | .008 | | .9853 | | | 84.4 | 91.3 |
| 1.42 | 1.109 | .020 | 58.6 | | 136.1 | 83.7 | | |
| 1.46 | 1.137 | .020 | 57.2 | | 135.3 | 81.7 | | |
| 1.43 | 1.117 | .025 | 56.0 | | 135.3 | 80.0 | | |
| 1.42 | 1.109 | .625 | 53.5 | | 133.7 | 76.4 | | |
| 1.43 | 1.121 | .625 | 53.0 | | 133.7 | 75.7 | | |
| 1.43 | 1.121 | 1.25 | 51.5 | | 133.1 | 73.6 | | |
| 1.43 | 1.121 | 2.5 | 51.1 | | 132.6 | 73.0 | | |
| 1.45 | 1.129 | 2.5 | | .9720 | | | 76.9 | 83.1 |
| 1.43 | 1.121 | 5.0 | 50.6 | | 132.1 | 72.3 | | |
| 1.43 | 1.118 | 5.0 | | .9700 | | | 75.8 | 82.0 |
| 0.13 | 0.103 | 0.007 | 75.7 | | 139.9 | | | |
| .14 | .110 | .007 | | .9952 | | | 89.8 | 97.3 |
| .13 | .103 | .026 | 69.5 | | 139.4 | 99.3 | | |
| .13 | .103 | .625 | 64.4 | | 136.3 | 92.0 | | |
| .13 | .103 | 5.0 | 61.3 | | 134.4 | 87.6 | | |

* Calculated from $\Delta H_f/\Delta H_\infty \times 100$, where $\Delta H_\infty = 70$ cal/g (2). † Calculated from $(v_a - v)/(v_a - v_c) \times 100$, where $v_a = 1.171$, $v_c = 0.9862$ (9), and $v = 1/\rho$ (ρ is the specific volume in cubic centimeters per gram, and the subscripts "a" and "c" refer to amorphous and crystalline forms, respectively). ‡ Calculated from $(v_a - v)/(v_a - v_c) \times 100$, where $v_a = 1.171$ and $v_c = 1.0002$ (10).

obtained from both methods were in excellent agreement. The thickness data in Tables 1 and 2 are from the calculated values.

Two aluminum pans which contained the polyethylene sandwiched between gold were put in the calorimeter, one on the reference holder and one on the sample holder. The composite sandwiches were heated up to 142°C under a reduced nitrogen pressure of 0.25 mm-Hg and a heating rate of 0.625°C/min. After being held at 142°C for 10 minutes, the polyethylene-gold composites were cooled to room temperature, under vacuum at different rates of cooling. No differences were found when samples were heated to 170°C prior to cooling. In some cases the same polymer sample was cycled at a variety of cooling schedules. The gold was dissolved from the polymer by suspending the gold-polyethylene-gold composite in a 3 percent solution of sodium cyanide for 7 to 10 days. The polymer films floated free of the gold foil. An x-ray fluorescence examination of the polymer indicated the absence of gold. The gold-free samples were heated at 10°C/min in nitrogen (30 ml/min), and the areas were determined by planimetry. The calorimeter was calibrated with pure indium after every determination.

The initial results of our investigation are shown in Tables 1, 2, and 3. It is clear that, for a gold substrate, the

Table 2. Change in the heat of fusion of gold-nucleated polyethylene, Marlex 6050, as a function of sample thickness.

| Film thickness (mil) | Weight of sample (mg) | Rate of cooling (°C/min) | Heat of fusion, ΔH_f (cal/g) | % Crystallinity θ_1^* |
|----------------------|-----------------------|--------------------------|--------------------------------------|------------------------------|
| 0.10 | 0.076 | 0.625 | 65.6 | 93.7 |
| .13 | .103 | .625 | 64.4 | 92.0 |
| .20 | .154 | .625 | 61.0 | 87.1 |
| 1.42 | 1.109 | .625 | 53.2 | 76.0 |
| 5.5 | 0.906 | .625 | 53.4 | 76.3 |
| 55.0 | 1.024 | .625 | 52.4 | 74.9 |
| 55.0 | 0.309 | .625 | 52.4 | 74.9 |
| Bulk | .100 | .625 | 53.6 | 76.6 |

* Calculated from $\Delta H_f/\Delta H_\infty \times 100$, where $\Delta H_\infty = 70$ cal/g (2).

Table 3. Density of gold-nucleated polyethylene (fraction of Marlex 6001 with an average molecular weight of 10,000).

| Film thickness (mil) | Weight of sample (mg) | Rate of cooling (°C/min) | Density at 23°C (g/cm ³) | % Crystallinity | |
|----------------------|-----------------------|--------------------------|--------------------------------------|-----------------|--------------------|
| | | | | θ_2^* | θ_3^\dagger |
| 0.12 | 0.092 | 0.0034 | 0.9962 | 90.5 | 97.9 |
| .09 | .072 | .0034 | .9998 | 92.4 | 100 |

* Calculated from $(v_a - v)/(v_a - v_c) \times 100$, where $v_a = 1.171$, $v_c = 0.9862$ (9), and $v = 1/\rho$. † Calculated from $(v_a - v)/(v_a - v_c) \times 100$, where $v_a = 1.171$ and $v_c = 1.0002$ (10).

rate of cooling and film thickness complement one another. Decreasing both the film thickness and the rate of cooling results in a polyethylene having a heat of fusion in excess of 70 cal/g and a melting temperature of $\sim 140^\circ\text{C}$. Although a value for ΔH_f of 75.7 cal/g is surprisingly high, it is not too unreasonable since Atkinson and Richardson (8) have reported recently that the heat of fusion of polyethylene at the melting point is 73.5 cal/g, about 5

percent above the highest previous estimate. Although the results we obtain are comparable to those for the high-pressure crystallized polyethylene, it is not clear as yet whether the highly crystalline polymer consists predominantly of the extended chain configuration.

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Multiple Genotypes in Individuals of *Claytonia virginica*

Abstract. *Supernumerary chromosomes are common among plants of Claytonia virginica found in a weedy population near the southwestern edge of its distribution. Roots, stems, and microsporocytes vary in chromosome number within the same plant in 68 percent of the population studied.*

Constancy of chromosome number within all individuals is merely a convenient fiction (1), for there is considerable evidence indicating that polyploidy and aneuploidy occur in various parts of many plants and animals (2). This largely ignored intra-individual variation in relation to cell, tissue, and organ differentiation needs critical exploration (3).

In November 1969, underground stems (corms) of *Claytonia virginica* L. were obtained from Carthage, Texas, and grown in the greenhouses of the Missouri Botanical Garden.

Twenty-two polyploid plants were studied from one population for both root mitosis from corms and microsporocyte meiosis, in addition to 12 plants previously examined (4). Once counts of chromosomes were made in these organs, floral stems were cut from each just below the paired floral leaves and planted in perlite under mist. A few developed adventitious roots from callus and these were studied for chromosome number when roots were about 1 cm long. Successful cuttings were potted and several plantlets with corms formed. Chromosome counts were made in cells from elongated roots as well as in the microsporocytes from the new floral stems.

Meiosis was largely regular with supernumerary chromosomes pairing normally [table 2 in (3)]. In mitotic plates supernumerary chromosomes appeared similar and indistinguishable from the characteristic complement of chromosomes which numbered 28.

Of the 23 plants from a total of 34 found to have multiple genotypes (68 percent), roots from corms averaged $2n = 28.3 \pm 0.4$ chromosomes per cell, roots from floral stems averaged $2n = 29.8 \pm 4.5$ chromosomes, and micro-

sporocytes averaged $2n = 29.5 \pm 0.8$ chromosomes (Tables 1 and 2).

Few supernumerary chromosomes were found in those roots originating annually from the corm—the basic genome of $2n = 28$ was found in 80 percent of all cells studied, and only 15 and 5 percent of cells had one or two extra chromosomes, respectively. This conservatism in multiple genotypic organisms appears more common for roots than for other organs in most species (5) although not universally so (6).

In contrast, supernumerary chromosomes are most frequent in meiotic cells of the anther. A total of 74 percent of cells had from one to five extra chromosomes. This high frequency is reflected by an average of 1.2 chromosomes more per cell than found in corm-originating root cells. Yet the average chromosome number of cells from adventitious roots arising from callus on floral stems is similar to the average found for microsporocytes, and not that seen in roots formed from corms, as might be expected. In fact, they averaged 1.5 chromosomes per cell higher with a wide range of $2n = 28-52$ and a large standard deviation. These adventitious roots, however, presumably arose from multiple tissues of an organ known or suspected to have great chromosomal variability (7), which is confirmed by the polyploidy found in this study. Even though a correlation exists between greatest cellular differentiation and greatest chromosomal variation, the number of cells having supernumerary chromosomes was only 47 percent, much less than that of the microsporocytes.

Two plantlets grew to sufficient maturity to be examined chromosomally. Results for one are given in Table 2. The second plantlet, propagated from a plant with $2n = 28$ (roots), 30, and 31 (microsporocytes), proved more complex. Its mature adventitious roots arising from a floral stem had $2n = 28, 29$, and 30 chromosomes (one root with each number), $2n = 31$ for microsporocytes from three different floral stems, and $2n = 25, 29, 33$, and 35 for microsporocytes from a single bud of a fourth floral stem. Vegetative reproduction has apparently been an inciting factor in genotypic variability for this individual.

Other correlations have been observed for those organisms with multiple genotypes. For example, a high number of supernumerary chromosomes in *Centaurea* is associated with dry climates (8), whereas individuals

Table 1. Multiple genotypes in roots from corms and microsporocytes of *Claytonia virginica*. Numbers in parentheses indicate number of roots and floral stem apices studied.

| Chromosome number in | | Plants (No.) |
|---------------------------|----------------------------|--------------|
| Roots from corms ($2n$) | Microsporocytes (as $2n$) | |
| 28 (11) | 29 (10) | 5 |
| 28 (11) | 30 (4) | 3 |
| 28 (6) | 31 (7) | 3 |
| 28 (1) | 30 (1), 31 (2) | 1* |
| 28 (3) | 31 (1), 33 (1) | 1 |
| 28 (3), 29 (1) | 28 (4) | 1 |
| 29 (1) | 28 (2) | 1 |

* Plus plantlet number 2 (see text).

Table 2. Multiple genotypes in roots from corms and floral stems and from microsporocytes of individual plants of *Claytonia virginica*. Numbers in parentheses indicate number of roots and floral stem apices studied. Each line is for one plant.

| Diploid chromosome number | | |
|---------------------------|-------------------------|------------------------|
| Roots from corms | Roots from floral stems | Microsporocytes* |
| 28 (2) | 28 (1) | 29 (2) |
| 28 (4) | 30 (2) | 30 (5) |
| 28 (2), 29 (2) | 28 (1) | 28 (5) |
| 28 (3), 29 (1) | 28 (2), 28 and 29 (1) † | 28 (2), 29 (1) |
| 28 (2), 29 (2) | 28 (1), 32 (1) | 28 (2) |
| 28 (1), 29 (3) | 28 (2) | 29 (3), 31 (1), 33 (1) |
| 28 (4), 29 (1) | 31 (1) | 29 (8), 31 (3) |
| 28 (3), 30 (4) | 28 (1), 30 (1), 31 (1) | 30 (1) |
| | 30 and 52 (1) † ‡ | |

* Chromosome number expressed as $2n$. † Two numbers in a single adventitious root (aneuploidy). ‡ Plus plantlet number 1 having $2n = 28$ in floral stem roots.