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**Electron Beam Irradiation of High
Density Polyethylene Pellets for
Thermal Energy Storage**

**Final Report of Task 1 and Task 2 for the
Period January 5, 1979 to January 4, 1980**

Joseph E. Davison, Ph.D.
Ival O. Salyer

OFFICE OF ADVANCED CONSERVATION TECHNOLOGIES

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PELLETS FOR THERMAL ENERGY STORAGE

Final Report of Task 1 and Task 2 for the Period
January 5, 1979 to January 4, 1980

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1. INTRODUCTION

The University of Dayton is conducting a program whose purpose is to investigate the crosslinking of high density polyethylene (HDPE) pellets by electron beam irradiation. The objective of the program is to define the conditions required to prepare thermally form stable high density polyethylene pellets by electron beam irradiation.

High density polyethylene has a melting point in the temperature interval of 125-140°Celsius and values for the latent heat of fusion have been reported as high as 50 calories per gram. At its melting point, the high density polyethylene changes from a white opaque crystalline solid to a clear transparent visco-elastic liquid phase. This property was especially useful in visual observations of the pellet bed during melt-freeze cycling of the thermal energy storage test column. The melting temperature of high density polyethylene makes it especially suited as a phase change thermal energy storage media for solar absorption air conditioning. Polyethylene melts and freezes reversibly and congruently. Thus the value for the heat of fusion remains at a constant value even after repeated cycling through the melting point.

The crosslinking of polyethylene pellets has a profound effect on their melting behavior. In their uncrosslinked condition, polyethylene pellets will melt in a normal manner, that is, the original shape of the pellets disappears as the pellets fuse together into one single mass in the liquid phase. After the pellets are crosslinked, they have the remarkable property of retaining their original geometric shape when heated above their melting point.

Since the pellets are thermally form stable, they may be utilized as a pellet bed in direct contact with the heat transfer fluid, whereas other phase change materials will require separate containers. In comparison to thermal storage media which utilize only the sensible heat of the material, polyethylene will utilize a much smaller volume and weight of storage media to deliver equal amounts of heat energy. Importantly, polyethylene can deliver this heat energy, which is stored in the change of phase, in a temperature interval between 125 to 140°Celsius.

In a previous investigation for the U.S. Department of Energy under Contract Number EY-76-C-05-5217, the crosslinking of polyethylene by chemical methods was investigated. The crosslinked polyethylene products were found to exhibit excellent thermal form stability and retained their initial value for the heat of fusion after more than 400 cycles through the melting point. These chemical methods are at least as effective in crosslinking polyethylene as the electron beam irradiation, but only at much higher costs per pound of finished product. The cost for preparing crosslinked polyethylene pellets by electron beam irradiation is estimated to add between one-half to one cent per pound to the cost of the polyethylene.

Although solar air conditioning provides an important application and supplied the initial impetus for this investigation, the properties of the thermal storage capability of HDPE and its melting temperature should also be considered for other thermal energy storage applications. For example, polyethylene should be considered for heating applications which will utilize the off-peak generating capacity and/or solar heating. A potential application for thermal energy storage systems exists in the load management of the electric utility industry. In this application they would utilize the excess off-peak electrical generation capacity to charge the thermal energy storage subsystem. During the peak hours for electrical generation, the thermal energy storage subsystem would be electrically disconnected and the thermal energy would be delivered from the stored heat energy. The economic incentives for this application already exists for those electric power consumers who have peak demand charges in addition to their normal energy charge. In this application, the consumer has the ability to level his own electric power demand and thus he is able to minimize his electric power demand charges.

The Dayton Power and Light Company has submitted to the Public Utilities Commission of Ohio a preliminary time-of-day rate schedule for its Residential, General Service, and Large Power rate groups. This proposed rate schedule will provide lower electric energy rates during the peak hours as compared to the present rate schedule. Thus, those consumers who have a need for heat energy could use thermal energy storage technology as a means to minimize their electric energy costs by charging their thermal energy storage subsystems during the off-peak hours and discharging them during the peak generation hours.

2. SUMMARY AND CONCLUSIONS

The overall objective of this project has been to define the electron beam irradiation conditions required to prepare thermally form stable high crystallinity polyethylene (HDPE) pellets which are suitable for thermal energy storage (TES) applications in the temperature interval of 120° to 140°Celsius. As originally proposed, the program consisted of three separate Tasks, only two of which were initially authorized and funded by DOE. The three Tasks were:

- Task 1. Define the optimum material and conditions for electron beam x-linking via evaluation of thermal form stability and retained heat of fusion of HDPE pellets in a laboratory (5 pound) TES unit.
- Task 2. Manufacture 250 pounds of crosslinked HDPE pellets under the optimum conditions defined in Task 1 and evaluate this TES material for stability to extensive thermocycling in a 250 pound "pilot plant" TES unit.
- Task 3. Manufacture 15,000 pounds of crosslinked HDPE pellets, evaluate this TES material for thermal form stability and retained heat of fusion in the 250 pound TES unit, and supply the remaining material to DOE for evaluation in one or more full scale home heating and cooling tests.

We have successfully completed the two funded tasks of this project.

We have conducted an evaluation of high density polyethylene pellets which were obtained from five different manufacturers. The values for the heat of fusion and the melting temperature were measured on all of the HDPE specimens received. Four different HDPE specimens were irradiated under different conditions of the total radiation dose received by the pellets, the electron beam accelerating potential, the electron beam current, the effect of inert atmospheres during irradiation processing, and the effect of stirring the HDPE pellets during the irradiation processing. The experimental values of the heat of fusion and the melting temperature of the irradiated HDPE pellets were measured and compared to the values of the as-received pellets to evaluate the effect of irradiation processing. The irradiated

pellets were heated to temperatures above their melting point for extended time intervals to determine their thermal form stability.

Thermal cycling tests were conducted on three of the HDPE samples wherein ethylene glycol was used as the heat transfer medium in a five pound thermal energy storage test unit. During these tests, the HDPE pellet beds were cycled through their melting points for more than 100 complete cycles. The temperature-time profiles of the pellet bed were monitored during these cycles to determine the performance of the pellets during the melting and freezing process. After completion of these tests, the pellet beds were removed and their thermal form stability was evaluated.

Following the successful completion of the laboratory evaluation of crosslinked HDPE pellets in the five pound laboratory TES unit (Task 1), the preparation and testing of larger scale quantities was undertaken and completed (Task 2).

Approximately 250 pounds of the selected HDPE (Alathon 7040) was crosslinked by electron beam irradiation under the optimum conditions already established (Task 1). Preliminary laboratory tests of thermal form stability, heat of fusion, and melting point were performed to confirm the suitability of the HDPE pellets for the larger scale tests. The pellets were then evaluated for stability to extended thermocycling (melt/freeze) in the 250 pound pilot plant TES unit. The temperature/time profile of the TES unit was then monitored through more than 100 successive cycles after which the unit was opened and visually examined. The pellet bed was found to be in excellent condition with little or no discoloration and the width of the open space between the pellets retained intact.

The net results of the Task 1 and 2 experiments lead us to the following conclusions.

1. The crosslinking of the HDPE is dependent primarily on the total radiation dose of the material and it is less dependent on either the accelerating potential of the electrons or the electron beam current.

2. The HDPE pellets irradiated to a dose of eight megarads have sufficient thermal form stability and retained heat of fusion to be utilized as a thermal energy storage material.

3. The melting temperature of the HDPE is not changed as a result of the irradiation processing.

4. The HDPE pellets can be satisfactorily irradiated in air. However, pellets irradiated in carbon dioxide or nitrogen atmospheres show less propensity to adhere together after being thermally cycled above their melting point.

5. The heat of fusion of the irradiated pellets shows a trend toward lower values with increasing radiation doses.

6. The irradiated pellets will adhere at their points of contact with each other in an unstirred pellet bed when heated above their melting point; in a stirred bed the pellets do not adhere together even when heated above their melting points for extended time intervals.

7. Pellets irradiated to a dose of eight megarads will maintain an open porous structure even after repeated thermal cycling through their melting point. Free flow of the heat transfer fluid was observed through the pellet bed after more than 100 cycles through the melting point of polyethylene.

8. The irradiated HDPE pellets can be in direct contact with the heat transfer fluid, ethylene glycol, and do not require separate containment.

9. A problem associated with the use of a commercial ethylene glycol automotive anti-freeze as the heat exchange medium, was the formation of a gelatinous silicate precipitate in the cool side heat exchanger. This precipitate was difficult to remove.

10. As a result of the successful completion of the first two tasks of this project, we recommend that Task 3 be implemented. The objective of Task 3 is the preparation of 15,000 pounds of electron beam crosslinked material, the evaluation of the crosslinked HDPE product on a laboratory and pilot plant size, and the implementation of a full scale application test.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1 Objectives of Tasks 1 and 2

The principal objective of the first two tasks of this project has been to define the electron beam irradiation conditions required to prepare thermally form stable high density polyethylene (HDPE) pellets. This objective has been achieved through the following experimental activities.

3.1.1 Objectives of Task 1

(1) Evaluate the heat of fusion and the melting temperature of several commercially available HDPE specimens as they are delivered by the manufacturer. It should be noted that HDPE products which are presently available have not been optimized with respect to their value of the heat of fusion. Values for the heat of fusion as high as 58 calories per gram, or even higher, may be possible in HDPE which is more completely crystallizable.

(2) Select HDPE specimens for electron beam irradiation exposures and irradiate the selected specimens to several different radiation dosage levels.

(3) Evaluate the irradiated HDPE specimens for their thermal form stability, retained heat of fusion, and their melting temperature.

(4) Select the irradiation conditions for which the HDPE specimens are thermally form stable and cycle the irradiated HDPE specimens through their melting point in a five pound thermal energy storage test column.

(5) Evaluate the performance of the HDPE specimens in the five pound thermal energy storage test column.

3.1.2 Objectives of Task 2

(1) Irradiate HDPE pellets for cyclic testing through the melting point in the 250 pound prototype thermal energy storage test unit.

(2) Evaluate the performance of the irradiated HDPE specimens in the 250 pound test unit.

3.2 Task 1 Experimental Results

3.2.1 Initial Evaluation of Commercial HDPE's and Selection of Candidate TES Materials

A total of 13 different commercially available high density polyethylene specimens were received from five different manufacturers. Listed in Table 1 are the names of the suppliers, the type of HDPE, and the nominal values of heat of fusion, and density. These were supplied in the form of pellets approximately 0.125 inches in diameter with the single exception of the Phillips Marlex TR-885 product which was supplied in both the pellet and powder forms.

3.2.1.1 Melting Point and Heat of Fusion of Commercial HDPE Materials

The values for the heat of fusion and the melting temperature were measured on each of the 13 HDPE specimens which had been received. These measurements were performed in a Perkin-Elmer Differential Scanning Calorimeter, Model DSC-2. The HDPE samples were contained in the standard Perkin-Elmer aluminum sample pans and a nitrogen atmosphere was maintained around the sample during the measurement. The HDPE samples were cycled between 25 and 150°Celsius in the scanning calorimeter at heating and cooling rates of 10°Celsius per minute. The scanning calorimeter was calibrated by measuring the area under the time-temperature scan of a measured quantity of high purity indium.

The melting and freezing behavior of the polyethylene specimens were observed to occur over a range of temperatures. The results of these measurements are presented in Table 2. The values of the temperatures T_1 , T_2 , and T_3 are the values, respectively, at which the onset melting transition is observed, (T_1 . The temperature at which the maximum displacement was observed in the time-temperature scan, (T_2), and the temperature at the end of the transition, (T_3). An analysis of the time-temperature scans showed that more than 90 percent of the heat of fusion occurred within $\pm 2^\circ$ Celsius of the temperature, T_2 , even though the heating and cooling rates were 10°Celsius per minute.

TABLE 1
HIGH DENSITY POLYETHYLENE SPECIMENS
RECEIVED FOR EVALUATION

<u>Supplier</u>	<u>High Density Polyethylene</u>	<u>Melt Index*</u> <u>(dg./min.)</u>	<u>Density*</u> <u>(g/cc)</u>
1. DuPont	Alathon 7040, Pellets	6.0	0.96
	Alathon 7050, Pellets	17.5	0.96
2. Phillips Petroleum	Marlex TR-885, Powder	30.0	0.964
	Marlex TR-885, Pellets	30.0	0.964
	6006, Pellets	0.7	0.958
	6030, Pellets	3.0	0.960
3. U. S. Industrial Chemicals Co.	LS-556, Pellets	8.0	0.955
	LS-606, Pellets	10.0	0.962
	LS-630, Pellets	32.0	0.962
4. Dow	42060N, Pellets	42.0	0.960
	12065, Pellets	0.9	0.965
	08064N, Pellets	8.0	0.964
5. Gulf	9606, Pellets		0.953

*nominal values supplied by vendor

TABLE 2
MELTING TEMPERATURE AND ENTHALPY OF
FUSION HIGH DENSITY POLYETHYLENE SPECIMENS

Sample	<u>Heating Cycle</u>				<u>Cooling Cycle</u>			
	Temp. (°K)			Enthalpy of fusion (cal/g)	Temp. (°K)			Enthalpy of fusion (cal/g)
	T ₁	T ₂	T ₃		T ₁	T ₂	T ₃	
7040	382	408	412	46.2	393	388	364	48.2
7050	385	406	410	47.3	393	387	370	44.2
TR-855, Powder	384	406	409	47.6	395	390	369	50.5
TR-855, Pellets	388	407	410	46.6	394	390	377	46.0
6006	384	411	420	47.1	398	392	365	49.1
6030	383	408	412	44.4	396	392	368	47.4
LS-556	385	406	410	38.2	393	390	377	41.2
LS-606	384	407	410	45.6	395	391	372	44.6
LS-630	381	407	411	47.3	394	390	367	45.6
8064	390	407	411	43.7	394	389	375	43.2
1-12065	387	407	411	40.5	395	390	370	42.2
4-2060	389	406	410	42.4	393	386	372	43.4
9606	370	405	410	45.6	394	389	360	43.3

3.2.1.2 Electron Beam Irradiation Conditions

Four of the commercial polyethylene products were selected for the electron beam irradiation experiments. These products were, the DuPont 7040 pellets, the Phillips TR-885 pellets, the U.S. Industrial Chemicals LS-630 pellets, and the Gulf 9606 pellets. The variables associated with the electron beam irradiation which were studied in our tests were:

1. The total irradiation dose received by the pellets,
2. The accelerating potential of the electron beam,
3. The electron beam current,
4. The effect of irradiation of the pellets in inert atmospheres, and
5. The effects of stirring the pellets during the irradiation processing.

The HDPE pellets were irradiated at the electron beam facility of the Radiation Dynamics Incorporated in Plainview, New York.

In the first set of tests, the HDPE pellet samples were irradiated to total dosages of 2, 4, 6, 8, 10, and 12 megarads. These irradiation tests were conducted with an electron beam accelerating potential of three million volts and at an electron beam current of 20 milliamperes. HDPE samples supplied by the four different manufacturers were included in these tests.

During the second set of tests, the HDPE samples were irradiated to a total dose of eight megarads. These tests were conducted at two different accelerating potentials, 1.5 and 4.5 million volts. In an additional test, the pellets received a total dose of 12 megarads at an accelerating potential of 1.5 million volts.

In the third set of tests, the HDPE samples were irradiated to a total dose of eight megarads at two different electron beam currents. The beam currents during these tests were 5.65 and 10.3 milliamperes. These tests were conducted in the three million volt electron beam facility.

The fourth set of irradiation tests were performed in several different gaseous atmospheres. The gases used were carbon dioxide, nitrogen, and helium. All of the HDPE samples in these tests were irradiated to a total dose of eight megarads. Two tests were conducted under the carbon dioxide atmosphere. In one test, the HDPE pellets were given a pre-irradiation heat treatment at 100°Celsius for 12 hours in the carbon dioxide atmosphere. In the other test, they were irradiated without the benefit of the pre-irradiation heat treatment.

The fifth set of tests were performed to determine the effect of stirring the pellets during the irradiation processing. These tests were performed only on the DuPont 7040 HDPE pellets. These pellets were irradiated to the eight megarad level.

3.2.2 Evaluation of Irradiated HDPE TES Pellets for Thermal Form Stability

The thermal form stability of the irradiated pellets was evaluated by placing a small quantity of the pellets, approximately 30 grams, in refluxing ethylene glycol, Prestone II type antifreeze. The pellets were maintained in the ethylene glycol at temperatures between 145° and 165° Celsius for extended time intervals. All of the HDPE pellets were observed to be a white opaque solid at room temperature and a clear transparent phase above their melting point. In all of these tests a finely divided white residue was observed to be present at the bottom of the flask upon the conclusion of the high temperature exposure.

3.2.2.1 DuPont Alathon 7040 HDPE Pellets

The condition of the DuPont Alathon 7040 HDPE pellets after the high temperature exposures in the ethylene glycol are shown in Figures 1a through 1m. The pellets in the as-received condition had fused into a single mass in less than two hours at 165°Celsius as shown in Figure 1a. Those pellets which had received a total dose of 2 and 4 megarads are shown in Figures 1b and 1c, respectively. Virtually all of the pellets had fused in a single mass within a two and one half hour time interval. Although the original shape of the pellets is still evident, individual pellets could not be easily broken off of the mass. The pellets which had received a total dose of 6 megarads had adhered together in a single mass after a 72

hour high temperature exposure in the ethylene glycol, but crumbled easily into individual pellets as shown in Figure 1d. At higher levels of radiation, 8, 10, and 12 megarads, the pellets remained free flowing, with only a few pellets adhering together as shown in Figures 1e, 1f, and 1g. Those pellets which were irradiated to a dose of 8 megarads, but at the lower electron beam currents of 5.65 and 10.30 milliamperes, had adhered together at their points of contact. These clumps could be easily crumbled into individual pellets as shown in Figures 1h and 1i. The pellets which had been stirred during the radiation processing remained free flowing after a 96 hour high temperature exposure in boiling ethylene glycol as shown in Figure 1j. These pellets had received a total radiation dose of 8 megarads. Pellets were also irradiated to the eight megarad level in gaseous atmospheres of carbon dioxide and of nitrogen. After a three hour exposure at 150°Celsius in ethylene glycol, the pellets were examined. The results are shown in Figures 1k and 1l. In both of these tests, some of the pellets appeared to have melted and fused together. In comparison to the other tests which had been conducted, the results of these last two tests were attributed to nonuniformity during the irradiation processing. All of these tests were conducted with an electron beam accelerating potential of 3.0 Million electron Volts (MeV).

In a separate test the DuPont pellets were irradiated to a dose of eight megarads with an electron beam accelerating potential of 1.5 MeV. These pellets were heated in ethylene glycol for two hours at 160°Celsius. As shown in Figure 1m, these pellets were essentially free flowing after the exposure, although some pellet clusters did form. These clusters could be easily fractured into individual pellets.

3.2.2.2 Phillips Marlex TR-885 HDPE Pellets

The condition of the Phillips TR-885 HDPE pellets after exposures in boiling ethylene glycol are shown in Figures 2a through 2i. The pellets in the as-received condition and those which received a radiation dose of 2 megarads had fused into a single mass in less than three hours at 165°Celsius as shown in Figures 2a and 2b. The original pellet shape cannot even be observed in these specimens. The pellets which had received total radiation dose of four and six megarads had formed a single mass with the original shape of the pellets clearly distinguishable as shown in Figures 2c and 2d. These pellets had been heated in ethylene

glycol to 165°Celsius for less than two hours. The pellets which had received a total dose of eight megarads had adhered at their points of contact after a three hour exposure at 165°Celsius. Individual pellets could be readily broken off after the specimen was cooled to room temperature as shown in Figure 2e. The pellets which had received a total radiation dose of 10 and 12 megarads showed a tendency to adhere together in small clusters after a 72 hour exposure at 165°C, but could be readily separated into individual pellets as shown in Figures 2f and 2g.

Two irradiation exposures of the pellets were conducted at electron accelerating potentials of 1.5 MeV and 4.5 MeV. The pellets irradiated with the 1.5 MeV electron beam received a total dose of 12 megarads, and those pellets irradiated with the 4.5 MeV electron beam received a total dose of eight megarads. The irradiated pellets were placed in ethylene glycol at 160°Celsius for two hours. The condition of the pellets after this exposure is shown in Figures 2h and 2i. Although the pellets had adhered together, they could be easily fractured into individual pellets.

3.2.2.3 U.S. Industrial Chemicals Company LS-630 HDPE Pellets

The conditions of the U.S. Industrial Chemicals Company LS-630 pellets are presented in Figures 3a through 3g. The HDPE pellets in the as-received condition and those irradiated to a total dose of 2 and 4 megarads fused into a single mass in less than one and one half hours in the boiling ethylene glycol as shown in Figures 3a, 3b, and 3c, respectively. The HDPE pellets which were irradiated to levels of 6 and 8 megarads had adhered together as a single mass in less than three hours at 165°Celsius. Although the original shape of the pellets is discernible, individual pellets could not be easily broken off. These pellets, after being cooled to room temperature, are shown in Figures 3d and 3e. At the 10 and 12 megarad levels of radiation, the HDPE pellets adhered together into a single mass after a 72 hour exposure in boiling ethylene glycol. However, as shown in Figures 3f and 3g, the pellets could be readily separated at their points of contact.

3.2.2.4 Gulf 9606 HDPE Pellets

The conditions of the Gulf 9606 HDPE pellets are presented in Figures 4a through 4h. As shown in Figure 4a, the as-received pellets had fused into a single piece within a two hour exposure in the hot

ethylene glycol. The pellets which received a dose of two megarads had adhered at their points of contact in a single piece but could be easily fractured at their points of contact into individual pellets. This sample is shown in Figure 4b. In Figure 4c is shown the sample which had received a four megarad dose. After two hours in boiling ethylene glycol, the pellets were still free flowing. In Figures 4d and 4e are shown samples which had received a six megarad dose. Although the pellets were free flowing after a two hour exposure in ethylene glycol at 160°Celsius, after a 96 hour exposure about one third of the pellets had adhered together. These clusters could be easily separated into individual pellets. At the 8, 10, and 12 megarad levels the pellets only showed a weak tendency to adhere at their points of contact and could be readily separated into the original pellets as shown in Figures 4f, 4g, and 4h.

A second set of six tests were performed on the Gulf 9606 HDPE pellets. The six specimens represented six different processing conditions. These conditions were:

1. The pellets were irradiated in air at room temperature.
2. The pellets were irradiated in air at 100°Celsius.
3. The pellets were irradiated in a nitrogen atmosphere at room temperature.
4. The pellets were irradiated in a nitrogen atmosphere at 100°Celsius.
5. The pellets were irradiated in an evacuated chamber at a pressure of 0.10 Torr.
6. The pellets were irradiated in an evacuated chamber after being outgassed at a pressure of 0.010 Torr.

The condition of the pellets was examined after exposures in ethylene glycol at 150°Celsius for a four to six hour time interval. The results of these tests are presented in Figures 5a through 5f. Of the six different processing conditions the least amount of adhesion among the pellets was observed for those irradiated under a nitrogen atmosphere at 100°Celsius.

3.2.3 Evaluation of Irradiated HDPE TES Pellets for Melting Point and Heat of Fusion

The values of the melting temperature and the heat of fusion were conducted on samples of the pellets after their irradiation treatment. These values were measured in the Perkin-Elmer Differential Scanning

Calorimeter, Model DSC-2. The results of these measurements are presented in Table 3. The results were compared to those values which were measured on the HDPE pellets prior to their irradiation. No significant difference was observed in the values of the melting temperature as a function of the radiation dosage. All of the irradiated pellets show that the liquid-solid transformation is still present in their irradiated condition. The results for the values of the heat of fusion do show a trend toward lower values with increasing radiation doses. The values for the heat of fusion as a function of the total radiation dose for the DuPont 7040 HDPE pellets are shown in Figure 6.

3.2.4 Thermocycling (Melt/Freeze) Evaluation of Irradiated HDPE Pellets in Laboratory (Five Pound) TES Unit

The performance of the HDPE pellets during repeated melt-freeze cycling was evaluated initially in a five pound thermal energy storage test unit. The five pound TES test unit is shown in Figure 7. This TES test unit was fabricated under an earlier ERDA contract, Number EY-76-C-05-5159, and its operation is fully described in the final report entitled, Form Stable Crystalline Polymer Pellets for Thermal Energy Storage, and dated July 1977. This report is available from the National Technical Information Service. A block diagram of the TES test unit is shown in Figure 8. The TES test unit consists of four basic components, the storage chamber which holds the HDPE pellets, an electrical heater which heats the heat exchange fluid, a cooling unit for removing heat energy from the system, and a metering pump to control the flow of the heat exchange fluid.

3.2.4.1 Description of Laboratory (Five Pound) TES Unit

Since the TES unit and its operation are fully described elsewhere, only a brief description is included here. During the heating portion of the cycle, the flow control valves numbered 1, 3, and 5 are open and those numbered 2, 4, and 6 are closed. The heat transfer fluid, Prestone II, is circulated by the metering pump in a clockwise manner through the heater and the storage column. The Prestone II is heated and then introduced at the top of the storage column. Thus the melting of the HDPE pellets in the storage column begins at the top and proceeds downward through the pellet bed.

TABLE 3

THE ENTHALPY OF FUSION AND THE MELTING TEMPERATURE OF
HDPE PELLETS AS A FUNCTION OF ELECTRON BEAM IRRADIATION

Sample	Radiation Level megarad	Heating Cycle				Cooling Cycle			
		Temp. (°K)			Enthalpy of Fusion (cal/g)	Temp. (°K)			Enthalpy of Fusion (cal/g)
		T ₁	T ₂	T ₃		T ₁	T ₂	T ₃	
7040	as received	382	408	412	46.2	393	388	364	48.2
	2	373	408	412	42.8	393	388	362	40.5
	4	373	408	414	43.2	393	387	361	40.9
	6	371	406	411	39.9	393	387	362	38.8
	8	369	407	411	42.9	393	388	363	40.1
	10	373	405	410	40.4	393	387	361	40.0
	12	371	405	410	37.9	393	387	363	35.7
	8 ¹	373	406	412	38.0	393	387	368	36.6
	8 ²	373	406	412	39.8	392	386	363	38.8
	8 ³	373	404	409	37.9	393	387	364	36.4
TR-885	as received	388	407	410	46.6	394	390	377	46.0
	2	376	406	409	48.7	394	390	360	48.8
	4	377	406	409	48.7	394	390	362	46.7
	6	373	407	410	51.0	394	390	360	50.2
	8	373	406	411	40.7	392	388	360	40.5
	10	373	406	411	40.2	393	388	362	40.2
	12	373	405	411	39.7	393	388	364	41.5
LS-630	as received	381	407	411	47.3	394	390	367	45.6
	2	372	405	408	45.4	394	390	361	48.3
	4	373	406	409	47.8	393	389	361	45.3
	6	373	406	410	49.0	393	390	360	46.2
	8	373	406	411	43.6	392	390	360	40.4
	10	372	406	411	42.0	392	388	359	40.7
	12	373	405	409	42.9	393	390	360	38.9
Gulf 9606	as received	370	405	410	45.6	394	389	360	43.3
	2	369	405	409	41.1	393	389	361	39.4
	4	368	405	410	41.8	393	388	360	36.3
	6	371	404	409	35.2	392	387	360	35.0
	8	367	403	407	35.5	392	388	360	33.0
	10	366	401	406	35.7	393	387	360	31.7
	12	367	401	406	37.0	395	387	360	34.3

Note: ¹ electron beam current - 10.30 milliamperes
² electron beam current - 5.65 milliamperes
³ HDPE stirred during radiation exposure

When the temperature at the bottom outlet is greater than 135° Celsius, the cooling portion of the cycle is initiated. During the cooling portion of the cycle, valves 1, 3, and 5 are closed and valves 2, 4, and 6 are opened. The Prestone II circulates in a clockwise fashion with the Prestone II entering the storage column at the bottom and exiting at the top. The temperature of the water bath radiator is adjusted to maintain the Prestone II at 100° Celsius at the inlet of the storage column. The freezing of the HDPE pellet bed is initiated at the bottom of the column and proceeds to the top of the column. When the temperature at the outlet port at the top of the column drops to a value between 100° and 115° Celsius, the heating portion of the cycle is again initiated.

The storage chamber consists of a 4 inch diameter glass tube which is 19 inches long and is insulated to minimize thermal losses during the cycling experiments. Three viewing ports through the insulation of the storage column allowed the HDPE pellets to be visually observed during the tests. The temperatures of the system were monitored at the inlet and outlet ports which are located at the top and bottom of the storage column and at a point within the storage column which is located approximately 6 inches from the bottom outlet.

3.2.4.2 Experimental Results of Thermocycling Tests in Laboratory (Five Pound) TES Unit

The HDPE pellets which were evaluated in this test unit were the DuPont 7040 pellets, the U.S. Industrial Chemicals LS-630 pellets, and the Gulf 9606 pellets. A five pound quantity of the HDPE pellets were loaded into the storage column and the unit was filled with Prestone II as the heat transfer fluid. The HDPE pellet bed was thermally cycled between 100° and 145° Celsius through more than 100 complete cycles. The melting and freezing of the pellet bed was detected by the thermal arrest of the thermocouples which sensed the temperatures within the storage column and at the outlet port of the heat transfer fluid. The melting and freezing of the pellet bed was confirmed by visual observation of the pellet bed through the observation ports through the insulation of the storage column. A typical plot of the temperature versus time during a cooling cycle is presented in Figure 9. The temperatures measured at the inlet and the outlet

of the heat transfer fluid and within the storage column are shown as a function of time from the initiation of the cooling mode of the cycle. As shown in this figure the temperature of the heat transfer fluid drops rapidly to 100°Celsius. The temperatures within the pellet bed shows the thermal arrest as the pellet bed freezes at 126°Celsius. The temperature of the heat transfer fluid at the outlet remains essentially constant until the freezing of the pellet bed has been completed. The completion of the freezing of the HDPE pellet bed is observed by the rapid drop in temperature of the heat transfer fluid at the outlet of the storage column. The temperature-time plots of the cooling portion of the cycle after more than 100 cycles were compared to the temperature-time plots which were measured before the accumulation of the heating and cooling cycles. This comparison of the performance of the HDPE pellet bed did not show any significant difference. The condition of the pellets after their removal from the storage column is shown in Figures 10a, 10b, and 10c.

Although the pellets had adhered to one another at their points of contact, the pellet bed maintained an open porous structure throughout the thermal cycling tests. Although the pellets adhered to one another in this test, the bonding between the pellets is very small. This observation is based on an experiment which was conducted on a fine quantity of the HDPE pellets after they were recovered from the thermal cycling tests. The clumps of HDPE pellets were heated in a stirred ethylene glycol bath to 150°Celsius. All of the clumps of pellets reverted back to individual pellets within a 30 minute time interval.

3.3 Task 2 Experimental Results

3.3.1 Election Beam Irradiation Crosslinking of 250 Pounds of HDPE-TES Pellets

Laboratory evaluation of five pound samples of HDPE-TES pellets, that were crosslinked by E-beam irradiation under various conditions led to the selection of "optimum" conditions for achieving thermal form stability consistent with minimum reduction in heat of fusion. These conditions were as follows:

• Accelerating potential	3,000,000 volts
• Current	20 milliamps
• Total dose	8 megarads
• Number of passes under beam	4
• Dose per pass	2 megarad
• Pellet agitation	Stirred after each pass
• Environment	N ₂ , CO ₂ , Air

In the small scale tests, irradiation in an inert environment of CO₂ or nitrogen (to exclude oxygen and concurrent ozone formation) appeared to significantly improve pellet thermal form stability (reduce surface adhesion) at a total dosage of eight megarads. Therefore, the first two samples of HDPE-TES pellets processed on the larger pilot plant (250 pound) scale included E-beam irradiation, at the Radiation Dynamics plant under these two inert atmospheres, respectively. The HDPE pellets (Alathon 7040) were inclosed, in ~20 pound lots, in 5 mil bags of low density polyethylene film. One bag was then placed in each 4 ft x 4 ft conveyer tray, purged with CO₂ (or N₂), sealed off, and conveyed (4 passes) under the E-beam. The pellets (inside the bags) were tumbled after each two megarad pass under the beam.

However, under these conditions, complete exclusion of air could not be quantitatively achieved, since a distinct ozone odor was noted in the irradiated pellets after the last pass. Additionally, it is believed that the heavy walled polyethylene container bags probably also detracted significantly from the uniformity and total dosage received by the pellets. These suppositions appeared to be verified when small scale lab tests showed the pellets to have non-uniform and inferior thermal form stability. Therefore, no large scale tests were done on these N₂, CO₂ samples since they would not be stable to thermocycling in the pilot plant TES unit.

Instead, a third 250 pound lot of Alathon 7040 HDPE-TES pellets were irradiated to a total dose of eight megarads in air (no polyethylene bag containers), with stirring between each two megarad pass. Additionally, precautions were taken to carefully spread the pellets in a thin uniform layer in the 4 ft x 4 ft container trays. Since the pellets were directly exposed to air, any ozone generated could be readily dissipated during the "stirring" after each pass. In contrast to the "bagged" N_2 , CO_2 atmosphere samples prepared previously, no ozone odor was noted by Radiation Dynamics personnel when the final product pellets were transferred into drums for shipment to Dayton.

The Alathon 7040 HDPE pellets irradiated in "air" as described above were shipped by truck (14 days in transit) from Melville, Long Island to Dayton, Ohio. Tests for thermal form stability and stability to thermocycling in the 250 pound pilot plant reactor were then performed as described below.

Importantly, processing in a completely inert atmosphere may still be advantageous to improved thermal form stability and should be further evaluated in any follow-on program. However, if such an inert atmosphere is to be effective, means must be provided to insure complete exclusion of oxygen. The polyethylene bagging method (only technique available at Radiation Dynamics at this time) is obviously unsatisfactory.

3.3.2 Thermocycling (Melt/Freeze) Evaluation of Irradiated HDPE Pellets in 250 Pound Pilot Plant TES Unit

3.3.2.1 Description of 250 Pound Pilot Plant TES Unit

As a result of the successful demonstration of the HDPE pellet bed in the five pound TES test unit, melt-freeze cycling

tests on a HDPE pellet bed were performed in a 250 pound prototype TES test unit. The 250 pound TES test unit is shown in Figure 11. This unit and its operation are fully discussed in the Final Report cited earlier, and only a brief description is included here. The flow diagram for the 250 pound unit is similar to the five pound test unit shown in Figure 8. The principle differences between the two units are:

1. The elimination of the water bath radiator in the 250 pound unit.
2. The incorporation of a water cooled heat exchanger in the line between the valves numbered 2 and 6 in the 250 pound unit.
3. The incorporation of an electrically operated control system which automatically controls the heating and cooling portion of the cycles.

The operation of the 250 pound unit is initiated in the heating mode with the valves numbered 1, 3, and 5 opened, and those numbered 2, 4, and 6 closed. The flow of the Prestone II is in a clockwise fashion. When the temperature of the thermocouple labeled T3 in Figure 8 shows a preset value greater than 135°Celsius, the cooling mode is automatically entered. The electrically operated valves 1, 3, and 5 are automatically closed and those numbered 2, 4, and 6 are opened. The flow of the Prestone II is also in a clockwise fashion during the cooling mode of the operation. The temperature of the fluid in the heat exchanger located in the line between the valves numbered 2 and 6 is maintained near 100°Celsius. In the cooling mode the heat transfer fluid enters at the bottom of the storage column and exits at the top. Thus, the freezing of the pellet bed is initiated at the bottom of the storage column and proceeds upward to the top. When the temperature of the heat transfer fluid at the exit of the storage column, as sensed by the thermocouple T4, drops below the preset value of 115°Celsius, the heating mode is automatically entered. All of the electrically operated valves change their position when the electrical heater is energized.

3.3.2.2 Experimental Results of Thermocycling Tests in 250 Pound Pilot Plant TES Unit

The storage column was filled with 250 pounds of the DuPont 7040 pellets. These pellets were irradiated to a dose of eight megarads at the electron beam facility of the Radiation Dynamics Incorporated. The flow of the heat transfer fluid through the storage column was channeled through the HDPE pellet bed by a series of baffles. Since the HDPE pellets have a tendency to adhere to one another at their points of contact, an aluminum screen with a mesh size of 20 openings per inch was used to partition the pellet bed into sizes which facilitate their easy removal at the end of the cyclic testing. In a separate test the HDPE pellets were observed not to adhere to the aluminum screen. The arrangement of the baffles and the aluminum screen is shown in Figure 12.

The HDPE pellet bed was tested through more than 100 complete heating and cooling cycles. The time interval required for heating the pellet bed through its melting point was approximately 75 minutes. The time interval required to cool the pellet bed was typically on the order of 30 minutes. The temperatures of the inlet, the outlet, and within the pellet bed are shown as a function of the time interval during the freezing portion of three cycles in Figures 13a, 13b, and 13c. The inlet temperature of the heat transfer fluid shows a slight decline over the time interval during the cooling portions of these cycles. The temperatures within the storage column and in the outlet of the storage column show a gradual decline over the major portion of the time interval during the cooling mode of cycle number 82 as shown in Figure 13a. The rapid decline of these temperatures near the end of the cycle indicate the completion of the freezing of the HDPE pellets. The isothermal character of the freezing of the HDPE pellet bed is readily observed on the cooling portions of cycle numbers 83 and 84, as shown in Figures 13b and 13c. During these cycles, the mass flow was maintained at a higher rate than in cycle number 82, and the temperatures of the pellet bed and the outlet drop rapidly over the initial portions of the cooling cycles. During the middle portion of the time intervals, the temperature within the pellet bed and in the outlet are reasonably constant. This characteristic is due to the isothermal freezing behavior of the HDPE pellet bed. Upon completion of the freezing of the pellet bed, these temperatures drop rapidly to the value of the temperature of the heat transfer fluid entering the storage column.

The HDPE pellet bed was removed and examined upon completion of 104 cycles. Views of the HDPE pellets after testing are shown in Figures 14a and 14b. As shown in these views, the pellets had adhered at their points of contact but had maintained an open porous structure which allowed the free flow of the heat transfer fluid through the pellet bed.

During the early portion of the melt-freeze cycling in the 250 pound TES test unit, a problem was observed during the cooling portion of the cycle. The pressure required to maintain an adequate flow of the heat transfer fluid was observed to increase during the cooling portion of the cycle. The value of the pressure was observed to increase with the number of cooling cycles. The pressure during the cooling portion of the cycle was approximately three times that required during the heating portion of the cycle.

After 30 cycles testing was suspended, and the valves, plumbing, and the heat exchanger were inspected. A deposit of a fine white powder was found to be present in the heat exchanger and the plumbing between the storage column and the heat exchanger. This powder was significantly more dense than the HDPE pellets. The powder was analyzed with the Energy Dispersive Analyzer for X-rays (EDAX) attachment of our scanning electron microscope. This analysis showed that the white powder contained a high concentration of silicon. The precipitate was attributed to the silicon-silicate additive present in the Prestone II and not to the HDPE pellets.

The heat exchanger and the plumbing line were flushed and the TES test unit was returned to operation. The pressure of the heat transfer fluid on the heating and cooling portions of the cycle were observed to be the same and the thermal cycling tests were continued and successfully completed with no further incident.

(a) Condition: as received
exposure interval: 2 hours

(b) Condition: 2 megarad dose
exposure interval: 2.5 hours

Figure 1. DuPont Alathon HDPE 7040 Pellets after Exposures in Ethylene Glycol. Temperatures of the Exposure were at 165° Celsius except as noted.

(c) Condition: 4 megarad dose
exposure interval: 2 hours

(d) Condition: 6 megarad dose
exposure interval: 72 hours

Figure 1. DuPont Alathon HDPE 7040 Pellets after Exposures in Ethylene Glycol. Temperatures of the Exposure were at 165° Celsius except as noted.

(e) Condition: 8 megarad dose
exposure interval: 72 hours

(f) Condition: 10 megarad dose
exposure interval: 96 hours

Figure 1. DuPont Alathon HDPE 7040 Pellets after Exposures in Ethylene Glycol. Temperatures of the Exposure were at 165° Celsius except as noted.

(g) Condition: 12 megarad dose
exposure interval: 96 hours

(h) Condition: 8 megarad dose, electron beam current
5.65 milliamperes
exposure interval: 3 hours

Figure 1. DuPont Alathon HDPE 7040 Pellets after Exposures in Ethylene Glycol. Temperatures of the Exposure were at 165° Celsius except as noted.

(i) Condition: 8 megarad dose, electron beam current
10.30 milliamperes
exposure interval: 2.5 hours

(j) Condition: 8 megarad dose. These pellets were stirred
after each 2 megarad exposure in the electron
beam.
exposure interval: 96 hours

Figure 1. DuPont Alathon HDPE 7040 Pellets after Exposures in Ethylene Glycol. Temperatures of the Exposure were at 165° Celsius except as noted.

(k) Condition: 8 megarads. These pellets were irradiated under a nitrogen atmosphere.
exposure interval: 5 hours at 150° Celsius

(l) Condition: 8 megarads. These pellets were irradiated under a carbon dioxide atmosphere.
exposure interval: 5 hours at 150° Celsius

Figure 1. DuPont Alathon HDPE 7040 Pellets after Exposures in Ethylene Glycol. Temperatures of the Exposure were at 165° Celsius except as noted.

(m) Condition: 8 megarad dose at 1.5 MeV
exposure interval: 2 hours at 160°Celsius

Figure 1. DuPont Alathon HDPE 7040 Pellets after Exposures in Ethylene Glycol. Temperatures of the Exposure were at 165° Celsius except as noted.

(a) Condition: as received
exposure interval: 3 hours

(b) Condition: 2 megarads
exposure interval: 3 hours

Figure 2. Phillips Marlex HDPE TR-885 Pellets after Exposures in Ethylene Glycol at 165° Celsius unless otherwise noted.

(c) Condition: 4 megarads
exposure interval: 2 hours

(d) Condition: 6 megarads
exposure interval: 2 hours

Figure 2. Phillips Marlex HDPE TR-885 Pellets after Exposures in Ethylene Glycol at 165° Celsius unless otherwise noted.

(e) Condition: 8 megarads
exposure interval: 3 hours

(f) Condition: 10 megarads
exposure interval: 72 hours

Figure 2. Phillips Marlex HDPE TR-885 Pellets after Exposures in Ethylene Glycol at 165° Celsius unless otherwise noted.

(g) Condition: 12 megarads
exposure interval: 72 hours

(h) Condition. 12 megarads. These pellets were irradiated with
an electron beam accelerating potential of 1.5 MeV.
exposure interval: 2 hours at 160° Celsius

Figure 2. Phillips Marlex HDPE TR-885 Pellets after Exposures in Ethylene Glycol at 165° Celsius unless otherwise noted.

- (i) Condition: 8 megarads. These pellets were irradiated with an electron beam accelerating potential of 4.5 MeV.
exposure interval: 2 hours at 160° Celsius

Figure 2. Phillips Marlex HDPE TR-885 Pellets after Exposures in Ethylene Glycol at 165° Celsius unless otherwise noted.

(a) Condition: as received
exposure interval: 1.5 hours

(b) Condition: 2 megarads
exposure interval: 1.5 hours

Figure 3. U.S. Industrial Chemicals Company HDPE LS-630 Pellets
after Exposures in Ethylene Glycol at 165° Celsius.

(c) Condition: 4 megarads
exposure interval: 1.5 hours

(d) Condition: 6 megarads
exposure interval: 3 hours

Figure 3. U.S. Industrial Chemicals Company HDPE LS-630 Pellets
after Exposures in Ethylene Glycol at 165° Celsius.

(e) Condition: 8 megarads
exposure interval: 3 hours

(f) Condition: 10 megarads
exposure interval: 72 hours

Figure 3. U.S. Industrial Chemicals Company HDPE LS-630 Pellets after Exposures in Ethylene Glycol at 165°C Celsius.

(g) Condition: 12 megarads
exposure interval: 72 hours

Figure 3. U.S. Industrial Chemicals Company HDPE LS-630 Pellets
after Exposures in Ethylene Glycol at 165° Celsius.

(a) Condition: as received
exposure interval: 2 hours

(b) Condition: 2 megarads
exposure interval: 2 hours

Figure 4. Gulf HDPE 9606 Pellets after Exposures in Ethylene Glycol at 160° Celsius.

(c) Condition: 4 megarads
exposure interval: 2 hours

(d) Condition: 6 megarads
exposure interval: 2 hours

Figure 4. Gulf HDPE 9606 Pellets after Exposures in Ethylene Glycol at 160° Celsius.

(e) Condition: 6 megarads
exposure interval: 96 hours

(f) Condition: 8 megarads
exposure interval: 96 hours

Figure 4. Gulf HDPE 9606 Pellets after Exposures in Ethylene Glycol at 160° Celsius.

(g) Condition: 10 megarads
exposure interval: 168 hours

(h) Condition: 12 megarads
exposure interval: 168 hours

Figure 4. Gulf HDPE 9606 Pellets after Exposures in Ethylene Glycol at 160° Celsius.

Figure 5a. Condition of Gulf 9606 Pellets after Exposure in Ethylene Glycol at 150°C for 4 Hours. These Pellets had been Irradiated in Air at Room Temperature.

Figure 5b. Condition of Gulf 9606 Pellets after Exposure in Ethylene Glycol at 150°C for 6 Hours. These Pellets had been Irradiated in Air at 100°C.

Figure 5c. Condition of Gulf 9606 Pellets after Exposure in Ethylene Glycol at 150°C for 4 Hours. These Pellets had been Irradiated in Nitrogen at Room Temperature.

Figure 5d. Condition of Gulf 9606 Pellets after Exposure in Ethylene Glycol at 150°C for 6 Hours. These Pellets had been Irradiated in Nitrogen at 100°C.

Figure 5e. Condition of Gulf 9606 Pellets after Exposure in Ethylene Glycol at 105°C for 4 Hours. These Pellets had been Irradiated in an Evacuated Chamber.

Figure 5f. Condition of Gulf 9606 Pellets after Exposure in Ethylene Glycol at 150°C for 6 Hours. These Pellets were Outgassed for 1 Hour at a Pressure between 0.010 and 0.013 Torr Prior to Irradiation in the Evacuated Chamber.

EFFECT OF ELECTRON BEAM IRRADIATION ON
THE HEAT OF FUSION OF DUPONT 7040 HDPE
(HEATING CYCLE)

Figure 6. The Values for the Heat of Fusion of DuPont Alathon 7040 HDPE Pellets as a Function of the Total Electron Beam Irradiation Dose.

Figure 7. View of the Five Pound Laboratory Thermal Energy Storage Test Unit.

Figure 8. Block Diagram of the Five Pound Laboratory Thermal Energy Storage Test Unit.

Figure 9. A Plot of the Temperatures at the Inlet and Outlet Ports of the Storage Column and within the Storage Column as a Function of the Time Interval on Cooling Cycle Number 112.

Figure 10a. View of the DuPont Alathon 7040 HDPE Pellets after being Thermally Cycled through their Melting Point in the Five Pound Thermal Energy Storage Test Unit.

Figure 10b. View of the U.S. Industrial Chemical Company LS-630 HDPE Pellets after Thermal Cycling Tests through their Melting Point in the Five Pound Laboratory Thermal Energy Storage Test Unit. (Note: The Aluminum Screen was used to Facilitate Removal of the Pellet Bed at the End of the Thermal Cycling Tests.)

Figure 10c. View of the Gulf 9606 HDPE Pellets after Thermal Cycling Tests through their Melting Point in the Five Pound Laboratory Thermal Energy Storage Test Unit.

Figure 11. View of the 250 Pound Prototype Thermal Energy Storage Test Unit.

Figure 12. View of the Baffles and Aluminum Screen Inserted into the Storage Column of the 250 Pound Prototype Thermal Energy Storage Test Unit.

Figure 13a. A Plot of the Temperatures at the Inlet and Outlet Ports of the Storage Column and within the Storage Column as a Function of the Time Interval on Cooling Cycle Number 82.

Figure 13b. A Plot of the Temperatures at the Inlet and Outlet Ports of the Storage Column and within the Storage Column as a Function of the Time Interval on Cooling Cycle Number 83.

Figure 13c. A Plot of the Temperature at the Inlet and Outlet Ports of the Storage Column and within the Storage Column as a Function of the Time Interval on Cooling Cycle Number 84.

Figure 14a. View of the Condition of the HDPE Pellet Bed after more than 100 Cycles in the 250 Pound Thermal Energy Storage Test Unit. Shown is the Condition of a Typical Layer of the Pellet Bed after Removal from the Storage Column.

Figure 14b. Close up View of the HDPE Pellet Bed Shown in Figure 14a. The Pellets Shown are within the Masked Area of Figure 14a. (Note: The HDPE Pellets have Retained their Physical Form after more than 100 Cycles through their Melting Point.)

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