# THE DEVELOPMENT OF A FREEZE-TOLERANT SOLAR WATER HEATER USING CROSSLINKED POLYETHYLENE AS A MATERIAL OF CONSTRUCTION

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### TABLE OF CONTENTS

	Page No
ABSTRACT	1
INTRODUCTION	2
OBJECTIVE	.5
PROCEDURE	6
RESULTS	1!3
DISCUSSION OF RESULTS	
Volume Expansion of Water During Freezing	16
Untested Parameters Affecting Freeze-Tolerance of XLPE Collectors	20
Formulation Parameters in Crosslinked Polyethylene	20
Design Parameters	22
Salts in the Water	25
Are 100 Freeze-Thaw Cycles Enough to Establish Freeze-Tolerance?	26
A Flat Spiral Coil of Tubing as a Collector Absorber Surface	. 28
Test Coil #10	29
Visco-Elastic Reaction of XLPE to the Strains of Repeated Freezing	30
The Performance of Carbon-Black-Reenforced XLPE Tubing as an Absorber for Solar Radiation	31
CONCLUSIONS	35
RECOMMENDATIONS	37
APPENDIX	38
BIBLIOGRAPHY	41

#### TABLES

Table No	<u>-</u>	Pa	age No	2
1.	Crosslinked Polyethylene Formulations used in Tubing Construction	·	8	
2.	Freeze-Thaw Test Results		14	

#### DRAWINGS AND PHOTOGRAPHS

Figure No.

5.

6.

#### Test Coil for Freeze-Thaw Test of Crosslinked Poly-1. 7 ethylene Tubing Freeze-Tolerant Solar Collector (Drawing) 11 2. Photograph of Freeze-Tolerant Solar Collector on the 3. 12 Test Rack at Los Alamos 4. Actual Performance of Freeze-Tolerant Collector compared to the Theoretical Performance of an Identical Collector 15 with a Collector Surface having an Absorptivity of 0.97

Failure of Tube Specimen #2

Failure of Tube Specimen #3

18

18

#### ABSTRACT

The feasibility of building a freeze-tolerant absorber for a solar water heater out of carbon-black-reenforced crosslinked polyethylene has been explored. Tenfoot tube specimens made from various crosslinked polyethylene formulations were filled with water at various pressures, and then placed into a deep freeze, then thawed and frozen again for 100 freeze-thaw cycles, or until the tube specimen failed. Tube diameters were measured before and after each freezing to determine how much distention the freezing caused, and how much permanent distention was caused by the strains of repeated freezings. Five tube specimens containing water at as high as 80 psi survived 100 freeze-thaw cycles.

Also, a flat plate collector was fabricated using as absorber surface a single 400 ft. tube of carbon-black-reenforced crosslinked polyethylene in the form of a flat spiral coil and this collector was tested for performance at the Los Alamos Scientific Laboratory. The performance test indicates that the absorbtivity of such a flat spiral coil to solar radiation is similar to typical black surfaces used on solar absorbers. Thus, it does seem very feasible that domestic water can be directly heated in a solar collector having an absorber made from crosslinked polyethylene, and that this collector can safely withstand at least 100 freeze-thaw cycles.

#### INTRODUCTION

As the price of energy continues to rise, the heating of domestic water by solar energy should be justified economically before other uses of solar energy because there is a steady year-round demand for hot water, and the cost of inherently expensive solar collectors can be amortized over more BTU's per year than say in space heating or air conditioning applications of solar energy. Unfortunately, water freezes during winter nights so a non-freezing collector fluid must be used and then the heat must be transferred from the collector fluid to domestic water through a heat exchanger. As Francis de Winter has pointed out very quantitatively, the costs of both the heat exchanger and the degradation of heat associated with the temperature difference across the heat exchanger are very considerable penalties (1). In his present study for ERDA (2), Francis de Winter also points out that non-freezing collector fluids have problems associated with viscosity, heat capacity, toxicity, and corrosion which increase still further the cost of transferring heat from the collector fluid to the hot water. If the designer of a solar collector elects to avoid the above costs and designs the collector to heat the water directly by the sun, then the solar collector must be drained infallibly on cold nights, or the collector must be made of a material sufficiently resilient to absorb the strains which freezing water can create, yet also be able to contain water at domestic hot water pressure without ballooning or corroding, and must be able to withstand without melting, occasional no-load temperatures when the supply of water is accidentally cut off on a sunny day. Polyset, Inc. suggested in an unsolicited proposal to ERDA that carbon-blackreenforced crosslinked polyethylene (XLPE) can probably do all these things, and the present Contract No. E (11-1) 2956 - Modification No. M001 - Supplemental Agreement to Contract No. EY-76-C-02-2956 is the result.

The basis of Polyset's confidence in carbon-black-reenforced crosslinked polyethylene (XLPE) was the outstanding long term hydrostatic strength which in the form of pipe it exhibits at 60° and 95° Centigrade (3). Also, carbon -black-reenforced cross-linked polyethylene is used on a large scale (about 100,000,000 lbs./yr.) as jacketing and insulation by the wire and cable industry, because it exhibits superb resistance to solar (ultra-violet light) degradation, and because it does not melt when the wire becomes overloaded with electric current, yet it retains excellent resilience at artic temperatures.

The crosslinked polyethylene formulation used for making hot water pipe differs substantially from the formulations used by the wire and cable industry, but it seemed likely that a compromise formulation could be found which would indeed be a suitable material of construction for a <a href="Freeze-Tolerant Solar Collector">Freeze-Tolerant Solar Collector</a>.

Crosslinked polyethylene also costs far less per unit volume than do the metals that might be used in solar water absorbers. Its cost in terms of energy required for manufacture are also very much lower than the cost of metals on a unit volume basis as one can see from the following table:

	Truckload Price ¢/lb.	Specific Gravity	Energy Required BTU's/lb.	Volume Cost ¢/cu.in. BTU/cu. <b>in.</b>			
Crosslinked Polyethylene	43	1.05	30,000	1.63	1,140		
Steel	16	7.6	13,000	4.4	3,570		
Aluminum	53	2.7	79,000	5.2	7,700		
Copper	65	8.92	34, <b>0</b> 00	20.9	11,000		

A low money cost and a low energy cost will both be essential for any solar collector planned for large scale use.

#### OBJECTIVE OF THE PRESENT PROGRAM

The objective of the present program as stated in the ERDA contract is to examine the feasibility of using carbon-black-reenforced crosslinked polyethylene as a material for solar collector absorbers having properties of low cost, long life, and tolerance to repeated freezing of water within the absorber.

ERDA asked that the freeze tolerance of crosslinked polyethylene be tested in the form of short (10ft.) lengths of crosslinked polyethylene tubing filled with water at various pressures in the range of domestic hot water pressures. These specimens were to be subjected to 100 freeze-thaw cycles, or until they burst and failed.

ERDA also asked that a simple solar collector using crosslinked polyethylene tubing be built using crosslinked polyethylene tubing filled with water as the absorber surface, and that its performance be measured and compared to similar solar collectors. Therefore, a collector design utilizing a flat spiral coil using cross-

linked polyethylene tubing was decided upon as the simplest design to use in that it

involved no problem of headering many tubes.

#### PROCEDURE

Tubes approximately 0.3" O.D. were made from nine different formulations of carbon-black-reenforced crosslinked polyethylene shown on Table I.

Fifteen 10 ft. specimens of this tubing were selected. The wall thickness at each end of the tube was measured by a micrometer. Each specimen was capped on one end, and on the other end it was connected to a 1 ft. length of 25 mm O.D. pipe made from white translucent crosslinked polyethylene obtained from Europe - see Figure (1).

The other end of the crosslinked polyethylene pipe was connected to a pressure gauge and to a needle valve through which high-pressure nitrogen could be introduced.

The connections between the crosslinked polyethylene tubing and the pipe and the pressure gauge and the nitrogen inlet, etc. were conventional fittings for copper tubing and copper pipe.

The black tubing was completely filled with tap water up to a level about one quarter of the way up the translucent pipe so that the volume <u>shrinkage</u> of the nitrogen between room temperature and the 0°F temperature of the freeze test would balance the volume <u>expansion</u> of the water freezing and result in a minimum fluctuation of pressure during freezing and thawing.

The tap water had the following composition:

Na	18.0 ppm	$50_4$	19 ppm	Cu 1. 1 ppm
Ca	13.0 ppm	$NO_2$	1.4 ppm	
Mg	5.2 ppm	$NH_4$	0.0 ppm	
C1	30. ppm	Fe	0.5 ppm	
Mn	.08 ppm	Si	1.0 ppm	Hardness 54 ppm

The tubing was then wound into a coil of about 20" in diameter so that it could conveniently fit inside of the deep freeze.

FIGURE I

# TEST COILS FOR FREEZE-THAW TEST OF CROSSLINKED POLYETHYLENE TUBING

SCALE: APPROXIMATELY 1/4"= |"

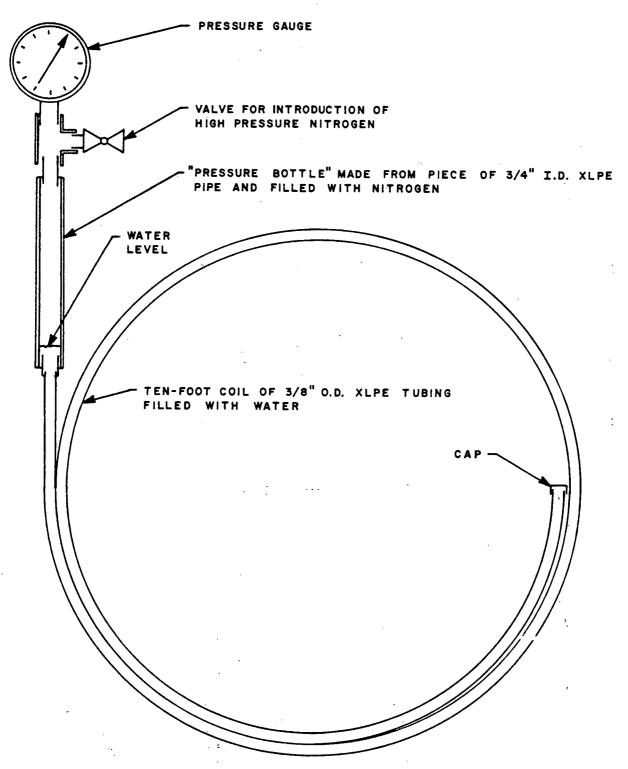


Table 1
Crosslinked Polyethylene Formulations Used in Tubing Construction

	Formulation Number	i	2	3	4	5	6	7	8	9
	Used in Test Coils Numbered	1	2,3,6&7	4	5	8	9	10,11,15	12	13&14
	Polyethylene Resin Density g/cc	.927	.96	.96	.96	.96	.96	.96	.933	.933
	Melt Index	2.17	3	3	3	3	3	3	2.33	2.33
I &	Loading of GPF* Carbon Black phr**	20	40.	20	40	40	40	20	40	· 40
~	Loading of AgeRite Resin D antioxidant phr	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
	Type of crosslinking agent	Lupersol 130	Same	Same	Same	Same	Same	Same	Varox	Lupersol 130
	Loading of crosslinking agent phr	2.5	2.5	2.5	4.0	1.5	4.0	1.5	1.5	1.5

<sup>\*</sup>GPF is General Purpose Furnace Black

<sup>\*\*</sup>phr is parts by weight per 100 parts by weight of resin.

In the initial tests the connection between the tubing and the pipe was insulated with Dacron padding in order to be sure that the water did not freeze inside this metallic connector before it froze inside of the XLPE tubing, and thereby trap water in this tubing during the freezing process, and subject the tubing to a freezing strain greater than what would occur in a solar collector. Later it was found that the tubing performed perfectly satisfactory without this Dacron insulation so it was removed. As mentioned above, the water in the tubing was subjected to hydrostatic pressure by means of bottled nitrogen fed through the needle valve.

The diameters of each tube were measured by micrometer at least two points on the tube, and at each point the diameter was taken twice, once in the plane of the coil, and once perpendicular to the plane of the coil.

The tube specimens were then placed in a deep freeze held at 0°F for at least three hours by which time the water inside was solidly frozen. Then the specimens were removed from the deep freeze, and the tube diameter immediately remeasured, taking care to measure these diameters at exactly the same points where the diameters were taken before freezing. The tube specimens were then allowed to thaw at room temperature or slightly above. (On cold days they were placed at the outlet of a space heater fan.)

After the water inside the test specimen had thoroughly thawed the tube diameters were again measured and then the specimens refrozen. This test procedure was continued for 100 freeze-thaw cycles, or until the tube specimens had failed from the strain of water freezing and swelling inside. During the first several freeze-thaw cycles the tube walls usually stretched irreversibly and acquired a permanent stretch of 1 to 4%. However, after twenty or thirty cycles the tube walls did not

continue to stretch irreversibly and the diameters before freezing seemed pretty constant. Thereafter, the diameters were measured less frequently. However, for the last five cycles, i. e. cycles 96 - 100, the tube diameters were once again measured before and after freezing.

Two solar collectors were then constructed. The first collector was fabricated from 403 ft. of 0.336" O.D., 0.047" wall thickness tubing made from Formulation Number 8 on Table 1. This tube was coiled into a square (42" x 42") flat spiral coil with the tubing bent on a 2" radius, so in theory there would be an area of 4" x 4", or 16 square inches, where sunlight would not fall upon a crosslinked polyethylene tube. Thus, theoretically less than 1% of the sunlight entering the collector has no black tubing to land on. The rest of the collector is pretty conventional as indicated in Figure (2).

This collector was sent to the Los Alamos Scientific Laboratory for performance tests. Figure (3) is a photograph of this collector on the test rack at Los Alamos. The second collector was fabricated from two 218 ft. lengths of 0.36" O.D. tubing made from Formulation Number 7 on Table 1. These two tubes were laid in parallel in the shape of a flat spiral coil, 43 3/8" x 43 3/8". The purpose of using two parallel tubes of about half the length of one tube is to reduce the pressure drop of the water flowing through the collector to a pressure down into the range that conventional hydronic heating system pumps can handle.

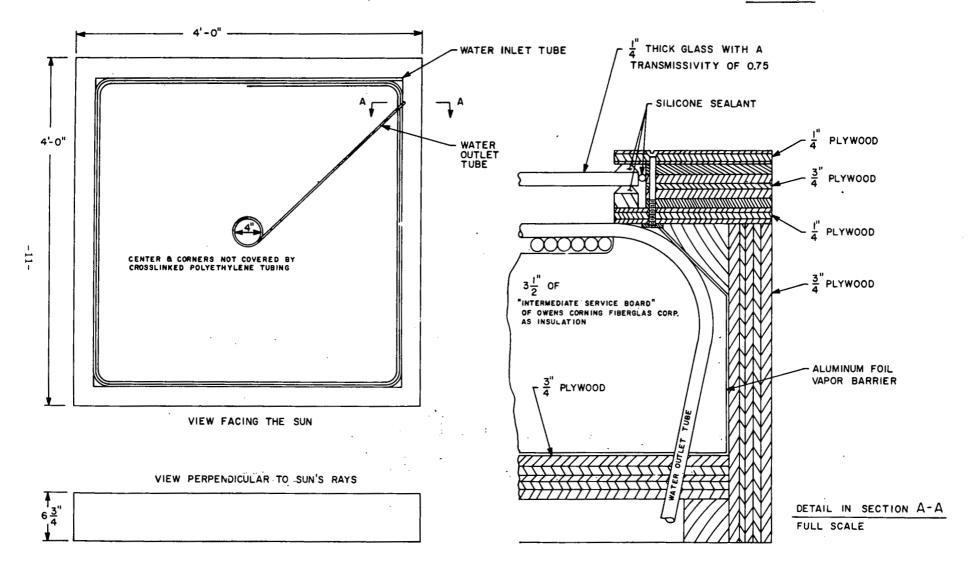
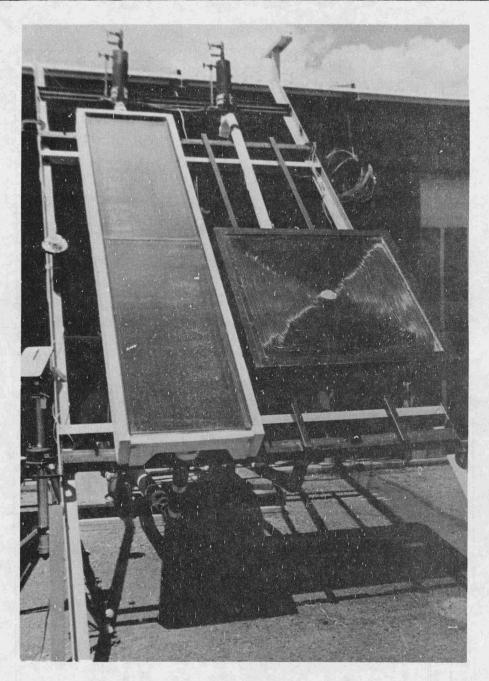


FIGURE 3

# Photograph of Freeze-Tolerant Solar Collector on the Test Rack at Los Alamos



#### RESULTS

The results of the freeze-thaw tests are shown on Table 2. Five of the fifteen test specimens survived one hundred freeze-thaw tests, and the others failed, after surviving somewhere between four and seventy-four freeze-thaw cycles. The results of the performance test are shown in Figure (4). The straight line is the performance "curve" of the collector fabricated from a flat spiral coil of crosslinked polyethylene tube. It is the "least squares" line of many data points taken in the range of  $T_F - T_A/I = 0$  to 0.4. The slightly curved line is the performance curve that should be expected from a conventional collector having the same area, the same single glazing (with a transmissivity of 0.75), and the same insulation, but having an absorber surface with 0.97 for both its absorptivity and its emissivity.

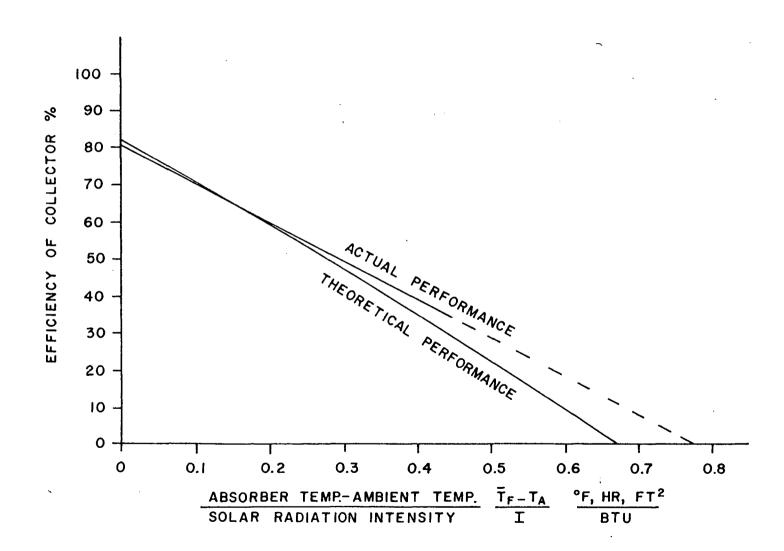
TABLE 2
FREEZE-THAW TEST RESULTS

	,	ı.	l					ı	1		,	ſ	1		i	1
Test Coil Number			2	3	4	5	6	7	8	9	10	11	12	13	14	15
Made from Formulation Number		1	2	2	3	4	2	2	5	6	7	7	. 8	9	9	7
Outside Diameter of Tube* Inches		0.390	.274	.282	.389	.397	.264	.271	.385	.385	.333	.367	.384	3405	.362	.359
Wall Thickness of Tube Inches		0.049	.016	.012	.021	.030	.020	.018	.046	.043	.023	023	.040	.035	.0353	.025
Water Pressure psi		30	52	53	40	40	30	40	56	80	52	82	56	1.15	94	131
Hoop Stress in Tube Wall psi		104	419	597	350	245	183	281	206	318	350	613	241	501	411	875
Average Percent Distention of Tube Diameter during/and After Freezing, cycles	1- 5	2.38/-	1.52/-	0.92/-	3.45/-	1.12/-	0.68/-	1.37/-	0.14/-	1.50/-	3.10/-	2.80/-	0.56/-	1.22/-	2.10/-	3.37/-
**	6- 10	2.35/0.43	-		3.33/0.30		0.06/1.55	1.67/0.63	0.96/(-0.22)	1.36/0.52	2.43/1.09	2.18/1.86	0.83/1.39	1.17/8.45	2.81/2.38	3.22/2.54
" 1	11- 15	1.17/1.38			2.98/0.02		(-0.33)/2.56	1.06/0.36	1.09/(-0.22)	1.24/0.93	2.86/0.89	2.87/2.98	1.75/2.93	1.52/8.90	3.44/4.29	3.36/2.48
" 1	16- 20	2.08/0.33					0.91/1.86		0.95/0.06	0.77/1.25	3.62/1.11	2.28/2.73	1.38/2.44		<b> </b>	
" 2	21- 25	2.14/(-0.28)					0.93/2.35		1.44/(-0.04)	0.75/1.38	3.31/0.73	2:92/3.12	1.76/2.63			
" 2	26- 30	2.13 (-0.31)			,		2.1/1.00		1.30/(-0.08)	1.17/1.09	3.65/1.04	3.10/3.60	0.79/2.44			
	31- 35	1.68/(-0.16)					1.55/1.35		1.08/0.03	1.73/0.81	3.91/1.62	2.62/4.13	2.08/2.40	•		
	36- 40	1.28/0.11	<u>s</u>	<u>s</u>			0.78/197	cycles	0.64/(-0.03)	0.59/1.04	4.29/1.56	2.57/3.87	2.45/2.50	cycles	<del>ភ</del>	cycles
	41- 45	1.39/0.03	Tube ballooned and failed after 4 freeze-thaw cycles	Tube ballooned and failed after 6 freeze-thaw cycles	cycles	cles	0.58/2.24	}	0.97/0.05	1.40/0.87	4.01/0.76	2.61/3.99	2.25/2.87		and failed after 12 freeze-thaw cycles	Š
	46- 50	2.22/(-0.36)	thaw	thaw.	17 cy	7 cycles	2.58/2.10	16 freeze-tha	0.95/(-0.34)			3.27/3.82	2.30/2.91	freeze-thaw	Ę.	freeze-thaw
" 5	51- 55	1.64/0.01	-əzə	-929		fter	3.27/1.42	28		1.71/0.66	4.92/0.27	3.11/3.75	<u> </u>	ž	8	97,34
>, <b>" - 5</b>	56- 60	2.25/0.03	4 fre	6 fre	ed a	led a	2.17/1.08	16 fr	0.62/(-0.35)				<u> </u>	<u> </u>	12 fr	13 fr
	61- 65	1.48/0.37	fter	fter	halt	hali		after	ļ	1.77/1.13		ļ <del></del>	1.07/3.34	after	نو	and failed after 13
· · · · · · · · · · · · · · · · · · ·	56- 70	1.42/0.01	eda	ed a	test	iest	<u>8</u> 8	ed a		·	4.75/1.01	3.29/3.28	<u> </u>		9 9	ed af
2	71- 75	1.87/0.08	l fail	j <u>e</u>	aks,	aks	failed	failed	0.97/(-0.10)		·——	failed		and failed	fail	fail
" 7	76- 80	1.84/0.33	anc	anc	ole le	ole le	and	anc			<del></del>	ξ Cλ			and	
" 8	81- 85	1.87/0.47	onec	oned	inhc	inh	oned eze-t	onec				than the		oued ou	] Je	pauc
8	86- 90	1.60/0.54	olle olle	allo	Excessive pinhole leaks, test halted after	Excessive pinhole leaks, test halted after	e ballooned and f	Tube ballooned and	·			) on 22		ballooned	Tube ballooned	Tube ballooned
•• 9	91- 95	1.76/0.13	be b	2 A	cess	cess	Tube b after 6	pe b				balle 74 fr		Tubeb	pe p	De D
" 9	96-100	2.03/0.45	₽.	구.	Ēx	Ex	Τ̈́u	1	1.81/0.31	1.36/1.22	4.38/0.37	Tube ballooned and after 74 freeze-thaw	1.43/3.25	-, 루	T.	2
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<sup>\*</sup>Based on the average of 20 measurements taken during the first 5 freeze-thaw cycles.

FIGURE 4

THE ACTUAL PERFORMANCE OF CROSSLINKED POLYETHYLENE COLLECTOR COMPARED TO THE THEORETICAL PERFORMANCE OF A CONVENTIONAL SOLAR COLLECTOR HAVING THE SAME GLAZING AND INSULATION AND AN ABSORBER SURFACE WITH A SOLAR ABSORBTIVITY EQUAL TO 0.97



#### DISCUSSION OF RESULTS

#### Volume Expansion of Water during Freezing

When pure water freezes at 32°F its volume expands from .01602 cubic feet per pound to a volume of .01747 cubic feet per pound. This is a 9.05% volume expansion, and if one assumes an equal expansion in all directions this would be a 2.93% linear expansion. Freezing of water inside of a tube does not take place instantaneously because a finite amount of time is required to remove the latent heat of fusion from Therefore, during the gradual volume expansion the unfrozen liquid water in the tube has a chance to escape, so long as its escape path does not itself become blocked by ice. If freezing takes place very slowly, and the escape route for unfrozen water is held open there should be no expansion of the tube during freezing. In fact, the dimension of the tube could actually contract due to thermal contraction. This appeared to be the case in the initial freezing cycles of Coil #6. After the first several freeze-thaw cycles it was decided to remove the thermal insulation from the outlet end of the 10 ft. coils so that they would freeze as rapidly as the rest of the tube (even more rapidly since the outlet fitting was made of brass and had a smaller diameter than the XLPE tube itself.) As a result a considerable amount of unfrozen water became trapped in the tube specimen. Then as this remaining unfrozen water did freeze and expand, it compelled the tube to expand with it.

More properly we should say that the ice, expanding into the trapped unfrozen water, subjected the entire tube length to an increasing internal pressure, thus imparting an increasing stress in the tube wall. Since no tube is perfectly uniform in wall thickness, some section of the tube would tend to distend under this increasing pressure more than others, and in so expanding would open up a bigger volume for

the yet unfrozen water. Thus, it would be the section of the tube that had the thinnest wall, and yielded the most to the increased pressure due to water freezing that would be subjected to the most extreme strains due to water freezing. This section with the thinnest wall would have to absorb the strain of the last bit of water freezing after it had already been distended more than any other section of the tube. This strain is far greater than the 2.93% strain that would occur if all of the ice froze instantaneously and expanded evenly in all directions. One can see in Figures (5) and (6) that in the region of failure the tube ballooned up to a diameter far greater than the average diameter. Therefore, it became clear early in the freeze-thaw test program that a distinctly more rubbery and resilient crosslinked polyethylene compound would be needed to absorb the actual strains of water freezing than had originally been anticipated. The early failure of Coils #2 and #3 indicated that Formulation 2 was too stiff and unyielding.

Rubbery or resilient crosslinked polyethylene compounds are not as strong as are the stiffer formulations. Nevertheless, they appear to be adequate to withstand domestic water pressure, yet tubes made from these more resilient formulations will not have to be so thick-walled that solar heat will have much difficulty in passing through the tube walls and into the water.

The thermal conductivity of crosslinked polyethylene is approximately 0.3 Fft.

Therefore, assuming a maximum solar heat flux of 300 BTU/hr./ft. the maximum temperature drop of the solar energy passing through the tube wall perpendicular to the solar rays can be calculated from the following equation:

300 = 
$$\frac{.3\eta}{t/12}$$
 T, where  $\eta$  = collector efficiency t = tube wall thickness in inches

The thickness of the tube wall needed to contain the internal hydrostatic pressure can be calculated from the following equation:  $\frac{H = p (D-t)}{2t}$ 

FIGURE 5
Failure of Tube Specimen #2

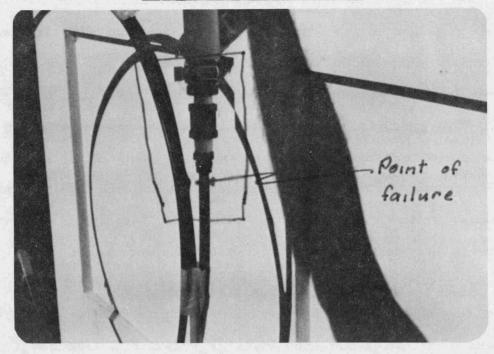
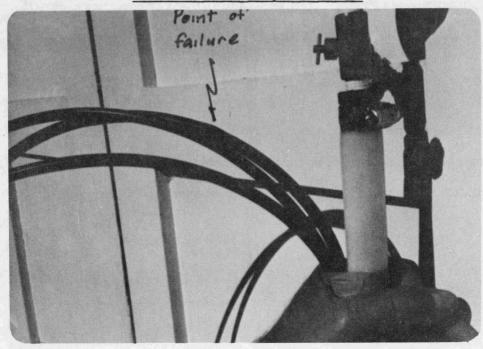


FIGURE 6

Failure of Tube Specimen #3



Where  $\underline{H}$  represents the design hoop stress\* of the tube wall material,  $\underline{p}$  equals the internal pressure,  $\underline{D}$  represents the outside diameter of the tube,  $\underline{t}$  equals the thickness of the tube wall in inches.

These two equations can be combined to give the following expression which permits one to calculate the maximum local temperature drop for heat flowing through the tubes as a function of the design hydrostatic pressure, the tube diameter, the collector efficiency, and the design hoop stress of the collector material.

$$T = \frac{8.33 \eta \, Dp}{2 \, H + p}$$

For instance, if we had a collector made up of 3/8" O.D. tubes based on Formulation #8, which can safely be subjected to a hoop stress of 241 psi for 100 freeze-thaw cycles, and if we assume that this collector would be operating at 32% thermal efficiency, (which would be typical for a double-glazed collector operating in an ambient temperature of 10°F while producing 150°F water during periods of pure sunshine radiating at 300 BTU's per hour per ft. 2) the maximum temperature drop across the tube wall, perpendicular to the sun's rays, would be 0.79°F. temperature drop would, of course, be lower at other points around the circumference of the tube because they would have less heat to transmit per unit area. It is worth noting that as the efficiency of the collector drops, so does the  $\Delta$ T across the tube wall. Thus, when the sun is weak or the outside air cool and the collector marginally active, the temperature drop across the tube can become very low indeed. On the other hand, if the outside air is warm and the sun is strong, there would be a much larger  $\Delta T$  across the tube wall, but during such periods the efficiency of the collector becomes high anyway, and a small drop in efficiency due to the \*Hoop stress is the maximum local stress imparted by the internal hydrostatic pressure to the tube wall.

temperature drop through the tube wall will be tolerable.

#### Untested Parameters Affecting Freeze-Tolerance of XLPE Collectors

There are many as yet inadequately-tested parameters in the formulation of crosslinked polyethylene and in the design of the collector which will affect the ability of the collector to withstand the strains of water at domestic water pressure freezing inside of it.

It may turn out that a freeze-tolerant solar water heater can easily be made without examining these many parameters. However, if service tests on cross-linked polyethylene collectors prove that an improvement in freeze tolerance or strength is needed then the following parameters should be investigated.

#### Formulation Parameters in Crosslinked Polyethylene

There are many polyethylene resins, and they are usually graded according to their density and their molecular weight, and also according to their content of non-hydrocarbon co-monomers. The present investigation did not include any study of the effect that the molecular weight of the polyethylene, nor of the effect of non-hydrocarbon co-monomers, have on the strength and resiliency characteristics of crosslinked polyethylene.

In the present investigation only two polyethylene densities were tried which is hardly an adequate exploration of that parameter since density of polyethylene homopolymer has a direct relationship to the crystallinity of the resin and therefore its strength and resilience.

Only one grade of carbon black was used and this at only two different loadings in the present investigation. These loadings were 20 parts/100 of resin, and 40 parts/100 There are many grades of carbon black and they are usually graded according to particle size, surface chemistry, and degree of aglomeration. We know that the

particle size of carbon black and its loading have a very pronounced effect on the strength and resiliency properties of crosslinked polyethylene. The degree of aglomeration and the surface chemistry of carbon black also to a lesser extent affect the resiliency and strength of crosslinked polyethylene. The amount of crosslinking agent used in a crosslinked polyethylene formulation also affects the strength and resiliency of the product, just as the amount of sulphur in a rubber compound affects the properties of the final vulkanizate.

Within limits, higher loadings of crosslinking agent do increase the resiliency and decrease the strength of a crosslinked polyethylene compound. However, the optimum amount of crosslinking agent is also governed by the amount and type of carbon black used in the formulation, and also by the molecular weight of the polyethylene.

Although the loading of crosslinking agent was varied in the present investigation this parameter was nevertheless inadequately explored.

Therefore, it is most unlikely that the combination of carbon black loading, carbon black type, and polyethylene resin type, and peroxide loading, selected in the present exploratory investigation will prove to be anywhere near the optimum for the construction of a freeze-tolerant solar water heater. Nevertheless, the indications are that formulations #5, #6, #7, #8, and #9, should be good enough to do the job. The fact that the two tubes, Coils #13 and #14, made from Formulation 9, failed after only 12 and 13 cycles should not be held against it, because these tubes were pressurized much more severely than was Coil #12 made from Formulation #8 which was identical to Formulation #9, except that different crosslinked agent was used. Both peroxide crosslinking agents can give excellent crosslinked products that are virtually indistinguishable in all respects if the other ingredients in the formulations are the same, and if each formulation is cured properly. In the curing process used

in the present investigation Lupersol 130 has some production advantages over Varox. The situation might be reversed if a different tube production process were used.

#### Design Parameters

There is reason to believe that the length-diameter ratio of a water tube has an effect on the amount of pressure that will build up inside the tube when the water freezes. The smaller the tube diameter and the longer the tube, the more difficult it is for the last bit of unfrozen water to escape, and thereby relieve the pressure inside the tube.

Freeze-thaw tests performed on 10 ft. specimens of <u>copper</u> tubing, 1/4" and 3/8" in diameter, which were performed during the course of the present investigation, support the theory that it is the last bit of unfrozen water that is trapped in the tube that does the real damage when it finally freezes.

These copper tube specimens were identical in length to the crosslinked polyethylene tube specimens, and were also capped on one end and filled with tap water, and frozen in the same deep freeze. However, these copper tubes were not pressurized at all - they were simply frozen at atmospheric pressure.

Diameter measurements were taken close to the open end of each copper tube and in the middle of each tube, and at a point close to the capped end of each tube.

Near the capped end and in the middle of each of these copper tubes the diameter distended irreversibly after each freezing, but at the open ends the diameters remained constant after repeated freezings. The average distention for each freezethaw cycle at the middle and capped end of each tube was only about 1.5% per cycle i.e. very much less than the 2.93% linear expansion of water upon freezing.

After five freeze-thaw cycles both tubes failed, one near the capped end, and one near the middle.

Since 1/4" copper tubing has a higher thickness-diameter ratio than has 3/8" copper tubing it can withstand a higher internal pressure before yielding; or in other words, it can subject the unfrozen water inside of it to a higher pressure, and thus be able to force this water through a narrower escape route than could the 3/8" copper tube. On the other hand, the 3/8" diameter copper tubes gave the unfrozen water a larger escape route. Therefore, the net result was that both tubes failed after the same number of cycles.

The 10ft. specimens of crosslinked polyethylene tubes used in the present investigation should give a reasonably good idea of the freeze tolerance of a collector made up of 10 ft. tubes where the pressure is vented at only one end of the collector, and should give an idea of the freeze tolerance of a collector made up of 20 ft. tubes where line pressure is maintained at both ends of the tube. (It seems unlikely that solar collectors would ever be designed where the water is shut into the collector by valves at both the inlet and outlet of the collector, because if a collector were ever exposed to the sun in this condition it would cause a "steam boiler explosion".)

Nevertheless, the 400 ft. coil of crosslinked polyethylene tubing in the collector made in the present investigation will, in its proposed service test, be subjected to a more severe freeze test than a 10 ft. tube specimen, because the unfrozen water during the freeze test will have a much greater distance to travel in order to escape.

On the other hand, the heat transfer rate from a given length of tubing in the solar collector during a service freeze test on a winter's night will certainly be far less than the heat transfer rate from the same length of tube in the freezethaw test of the present investigation because in the latter case the tubes were immersed suddenly into a 0°F deep freeze, whereas in a service test the temperature will not drop suddenly from above freezing to 0°F inside of the collector. The outside temperature does not drop that fast, and also the rate of heat transfer from the tubes will be greatly retarded by the glazings over the collector. Thus, the unfrozen water will have a greater time to escape in a service test, and this might conceivably counteract the extra pressure drop associated with the greater distance this water must travel in order to escape and relieve pressure. However, once the escape route for the unfrozen water is blocked by ice, the slow rate of freezing of the trapped unfrozen water inside the long flat spiral coil of the collector might greatly increase the distention of the tube at one concentrated point. If the trapped water is frozen very gradually it will have more time to migrate to the portion of the long tube which has the thinnest wall and which, therefore, yields the most to the ever-increasing pressure inside the tubing. If the weakest portion of the 400 ft. tubing can draw upon 40 times as much trapped water as occurred in the weakest point of a 10 ft. freeze-thaw specimen it may fail far far more readily than the 10 ft. specimen.

For this reason it should be very instructive to perform a service test on a parallel-tube XLPE collector at the same time and the same place as the service test on the flat spiral coil collector.

Since we do not know at this time whether the headers and the junctions of the headers and the parallel tubes can withstand the strains of freezing and thawing it would be necessary to insulate and heat trace the headers so that they will never be subjected to freezing.

As indicated on page 23 the distance between the headers could be as much as 20 ft. and still give the unfrozen water as much of a chance to escape as they did in the 10 ft. specimens that are capped on one end and used in the present investigation.

The heat losses from the heat-traced and insulated headers should presumably be very comparable to the heat losses from the similar lengths of the heat-traced and insulated water lines connecting the collector to the warm interior of the house.

#### Salts in the Water

Another unexplored parameter is the possible effect of various levels of dissolved salts that are typically found in drinking water.

I do not mean to suggest that the salt concentration is ever high enough to lower appreciably the freezing point of all of the water in the collector. However, I do suggest that whenever the freezing rate is slow (i.e. a freezing front advance of less than one inch per hour) these dissolved salts have time enough to diffuse ahead of the advancing freeze front and not become trapped between the ice crystals. (This fact was discovered during the development of freeze processes for desalinating sea water.) (4) Thus the salt concentration in the unfrozen water could increase many fold as the ice front approaches the center of the tube, and

this higher salt concentration could help some of the last bit of water to escape and relieve some of the internal pressure before this last remaining water becomes trapped and freezes and expands irresistably.

In most cases I doubt that the salt content of the water will have much effect on the freeze tolerance of a collector, but in areas where cold snaps are not very cold nor very sudden it might have an appreciable effect.

In any case, the very low dissolved solids content of the Manchester, Massachusetts, water (see page 6) used in the freeze-thaw tests, makes this water relatively easy to freeze totally, and, therefore, the freeze-thaw tests in the present investigation should give conservative results from the salt content point-of-view.

#### Are 100 Freeze-Thaw Cycles Enough to Establish Freeze Tolerance?

Whenever the repeated strains of freezing caused the XLPE tubes to distend permanently, the maximum permanent distention was usually achieved after 20 to 30 cycles. Thereafter, the diameter of the tubes distended a little more, but returned approximately to the same diameter that existed before the latest freezing. Therefore, it does seem unlikely that a collector made from crosslinked polyethylene will continue to grow after 100 freeze-thaw cycles, so failure from unending ballooning of the tubes also appears unlikely.

On the other hand, a failure did occur in test Coil #11 after 74 cycles - long after it appeared to have reached its stable diameter which was about 3% larger than the original diameter. Therefore, there may be a fatigue mechanism working here which cannot be detected by creep measurement.

The resistance to fatigue failure in metals can be determined fairly quickly by high speed flexing of a specimen. However, in a plastics material the stress-strain

characteristics are very time-dependent (also, very temperature-dependent), and it seems unlikely that a very much accelerated test could reliably establish the fatigue resistance of a plastics material such as crosslinked polyethylene, especially at the temperature of a tube wall surrounding water as it is about to become completely frozen. Extensive field tests appear to be the only way that freeze tolerance can be established with confidence.

Nevertheless, 100 freeze-thaw cycles should be more than adequate for the lifetime of solar collectors in Southern states. Four freezings per winter for twenty-five winters should be typical for a large portion of the United States because not all frosty nights are likely to be cold enough for a long enough period to freeze completely the contents of a solar collector.

Also, if the collector is drained most nights by an automatic drain valve, the collector's tolerance to 100 freeze-thaw cycles should amply take care of the inevitable malfunction of such an automatic drain valve sometime during the life of the collector. (It might be mentioned here that crosslinked polyethylene can very easily withstand the oxidative corrosion that occurs when air is introduced daily into the collector as a result of the nightly drainings, whereas metals find this kind of corrosion hard to take.)

Therefore, we can with some confidence say that the tests performed in the present investigation indicates that crosslinked polyethylene can be used as a material of construction for a non-drain freeze-tolerant solar collector suitable for all Southern states. We can also conclude that if a XLPE solar water heater is drained automatically on all but 100 nights during its lifetime by a fallible drain valve it can still be used in Northern states without bursting and failing.

#### A Flat Spiral Coil of Tubing as a Collector Absorber Surface

An absorber surface for a solar collector can be fashioned out of a tube by coiling this tube into a flat spiral. If just one tube is used there is no problem of joining a multiplicity of tubes into a header. Headers inherently involve problems of stress concentrations and other joining problems which could result in leaks, etc. For this reason, in the present investigation "to examine the <u>feasibility</u> of using - - - XLPE as a material for solar collectors having - - tolerance to repeated freezing of water within the absorber", the flat spiral coil design was adopted for the collector surface itself.

However, the flat spiral coil design has several disadvantages:

- (1) It cannot be drained easily.
- (2) It tends to have a much higher pressure drop than have the conventional parrallel-tube designs, and, therfore, a larger diameter tube must be used for a given pressure drop. This larger diameter in turn necessitates a proportionately thicker tube wall in order to withstand a given internal hydrostatic pressure. The thicker wall in turn results in a higher temperature drop through the tube wall for a given absorption rate of solar energy. A thicker wall also increases the raw material cost, and the thermal inertia of the collector.
- (3) During the period when the water inside of the collector is freezing, the unfrozen water has a greater distance to travel before it can escape from the collector and thereby relieve the internal pressure caused by the expanding newly-formed ice. This extra escape distance means greater pressure drop and therefore greater pressure on the tube wall.
- (4) During the no-load operation the water remaining in the collector will have a harder time to escape under the impulse of the expanding steam generated inside

of the collector. Therefore, dangerous vibrations, and possibly dangerous steam pressures might occur inside of the collector before all of the water is flushed out of it by the steam generated.

(5) When the perimeter of the flat spiral coil is to be rectangular or square, as the collectors made in the present investigation, the tubing in the coil is alternately straight when parallel to the side of the collector, and curved to make the bends at the corners. When a crosslinked polyethylene tube is bent it becomes slightly "egged" so its diameter in the plane of the coil becomes somewhat smaller than its diameter when it is not bent.

This reduced diameter gives solar light a chance to miss the absorber altogether and simply strike the insulation on the other side. This loss in projected area must be added to the loss in projected area resulting from the inability to coil the tube any smaller than a radius of 2" which in turn results in a 4" diameter hole in the absorber surface in the middle of the collector and for smaller segments in the corner.

In order to keep pressure drop within reasonable bounds the second collector was produced in the present investigation from two tubes, both wound parallel to each other into a flat spiral coil. In theory, this should reduce the pressure drop for a given flow of water by a factor of more than seven, assuming turbulent flow. The extra tube necessitated an extra connector at the inlet and outlet of the collector, but this was a modest additional cost compared to the savings in pumping power.

#### Test Coil #10

Test Coil #10 was unique in that the measured distention in tube diameter caused by freezing averaged 3 3/4%, or distinctly more than the 2.93% distention that one would predict from theory.

However, it should be remembered that the <u>tangential</u> stress in a tube wall is twice that in the <u>longitudinal</u> direction, and therefore in a tube made from a classical solid material which obeys Hooke's law, the tangential <u>strain</u> should be precisely twice as great as the longitudinal <u>strain</u>.

Furthermore, since plastics materials do not obey Hooke's law, but have convex stress-strain curves, a doubling of the stress more than doubles the strain.

Therefore, the 3 3/4% distention of the diameter is quite understandable.

Viscoelastic Response of Crosslinked Polyethylene to the Strains of Repeated Freezing.

In one respect the freeze-thaw tests performed in the present investigation were unrealistically severe. When any unoriented or isotropic plastics material is subjected to a stress, it does not return immediately to its original size after the stress is removed as does a piece of metal. For this reason one could not use an isotropic plastics material as a spring in a spring balance. Due to a "viscous" drag between the polymer molecules it takes time for these polymer molecules to move out from their most comfortable position when the plastics material is stressed, and when the stress is removed it takes time for these molecules to bounce themselves by thermal agitation back again into their most comfortable position.

During the freeze-thaw test of the present investigation the specimens were frozen and thawed sometimes twice a day. So the tubing really did not have much of an opportunity to return to its original diameter as it would have in actual service where it would be frozen once a day at most.

Furthermore, in actual service the tubing would usually be subjected to higher temperatures between freezings than they were in the freeze-thaw tests, so the thermal agitation bouncing the polymer molecules back into their original configuration would be stronger and less opposed by the viscous drag of the viscoelastic characteristic of the crosslinked polyethylene. Therefore, the 10 ft. test specimens were probably distended more during the freeze-thaw tests than would be the collector tubes of the same length and diameter in actual service.

# The Performance of Carbon-Black-Reenforced Crosslinked Polyethylene Tubing as an Absorber for Solar Radiation

The manufacturer (Libby Owens Ford) of the glass used in the collector tested at the Los Alamos Scientific Laboratory claims that the 1/4" thick untempered plate glass used in the collector has a transmissivity of  $75 - 1\frac{1}{2}\%$ . This glass is ordinary glass used for store windows, etc., and its low transmissivity can be attributed to its high iron content coupled with its 1/4" thickness. At first blush it seems impossible that the measured efficiency of the collector as shown in Figure (4) could ever be higher than the 75% transmissivity of the glazing, even when the collector temperature is the same as ambient and there are no heat losses (i.e. the conditions that occur at the "y" intercept on However, on further consideration one realizes that at "y" intercept conditions, the absorber surface receives not only shortwave solar radiation, but also a substantial amount of thermal radiation and convection from the glazing which it acquired by absorption of some of the solar radiation. Even at conditions indicated at points on the curve to the right of the "y" axis the efficiency is not as bad as one might expect because the solar heat absorbed in the glazing raises the temperature of this glazing above that which it would have if it had received its heat only by radiation and convection from the hot absorber surface. Therefore, the  $\Delta$  T between the absorber surface and the glazing is reduced, thereby reducing heat losses and increasing the collector's efficiency.

absorption and degradation of solar heat by the iron in the glazing is not a total loss for the collector's efficiency.

Since glass with untreated surfaces typically reflects 8% of the solar radiation striking it at near normal angles, we can calculate that approximately 17% of the solar energy is absorbed by the iron in the glass.

Of the 75% that is transmitted, about 1% missed the flat spiral coil of XLPE tubing and fell upon the insulation\* and was either reflected or absorbed and reradiated in all directions.

Of the 74% of the light transmitted that struck the flat spiral coil of the cross-linked polyethylene tubing presumably most of it was absorbed, but some was reflected.

Of the light (or <u>short-wave</u> radiation) that is reflected from the crosslinked polyethylene tubing, and from the insulation behind the polyethylene tubing back to the glass, 8% is again reflected and 17% absorbed. All of the heat radiating from the insulation is also absorbed by the glass. Therefore, distinctly more than 17% of the solar energy striking the outside of the glazing is absorbed by the iron impurities in this glazing, and if the absorber surface is maintained at the same temperature as ambient (i.e.  $T_f - T_a = 0$  on Figure (4) this solar heat absorbed in the glass is transferred in both directions by both radiation and convection. The amount which is transferred in each direction is

<sup>\*</sup>As indicated on page 10 the coil of XLPE tubing did not cover a 4" diameter circle in the center of the collector, nor four smaller segments in the corners of the collector. Also, the "egging" of the tube where it is bent leaves cracks between the tubes where sunlight can filter through.

governed to a great extent by how much wind is blowing outside of the glazing. If there is no wind the heat transferred in each direction would be nearly the same, and indeed the wind was very light during the period when performance tests were made on the crosslinked polyethylene collector at Los Alamos. The personnel at the Los Alamos Scientific Laboratory conclude that the performance curve of the crosslinked polyethylene collector submitted to them for tests had a 'y' intercept of 0.804 (i.e. when the collector surface was the same temperature as ambient the collector efficiency was 80.4%) and that this curve had a negative slope of 1.04.

Figure (4) also shows what the theoretical performance curve of a collector having an absorber surface with a solar absorptivity of 0.97 (typical for a good flat black paint), and a single glazing having a transmissivity of .75, and assuming that 8% of the solar energy striking this glazing is reflected, and 17% absorbed on the first pass that the solar energy makes through the collector. For this theoretical case the ambient temperature is assumed to be 80°F and no wind. One can see from Figure (4) that the performance of the crosslinked polyethylene collector is very close to what one might expect in theory from a conventional collector having the same single glazing with the rather low transmissivity of .75. At the time of this writing, September 21st, the Los Alamos Scientific Laboratory had not yet subjected the collector to anything approaching stagnation conditions, i. e. conditions approaching the points on the performance curve approaching the "x" axis.

Since radiant heat losses are not linear with temperature difference, but increased as a fourth power of the absolute temperature, the author predicts that

the slope of the performance curve will steepen from the minus 1.04, which best fit the present data points near the "y"axis, and that the actual measured performance curve will follow more closely the theoretical performance curve than the extrapolation of the present "least squares" line.

The Los Alamos personnel also noted that inadequate provision had been in the test collector for thermal expansion of the flat spiral coil so it buckled, and at some points actually touched the glazing. This should even further reduce the high temperature efficiency of the test collector.

The second collector which will hopefully be used for service tests at Colorado State University in a future ERDA contract has been finished with a glazing consisting of two panes of low-iron glass supplied by ASG industries. Presumably this glazing will substantially increase the efficiency of the collector when operating at domestic hot water temperatures on a cold winter day, and it will also make the collector far less affected by winter winds.

#### CONCLUSIONS

- (1) The objective of the present investigation has been achieved in that we can positively say that it is indeed more than <u>"feasible to use carbon-black-reenforced crosslinked polyethylene as a material for solar collector absorbers having - tolerance to repeated freezing of water within the absorber".</u>
- (2) A fairly resilient XLPE compound should be used in making a freeze-tolerant solar water heater. Formulations based on high density polyethylene should contain about 20 phr GPF carbon-black and low density resins about 40 phr GPF carbon-black, but the formulation parameters have by no means been optimized by the present investigation.
- (3) The pressure of water within a XLPE absorber during freezing can probably be held as high as that occurring in any likely domestic water supply without necessitating a construction so thick and heavy as to cause in normal operations more than one or two degrees Fahrenheit of temperature drop to transfer solar heat across the absorber surface into the water on the other side.
- (4) When XLPE tubing is bent into a square flat spiral coil, the cross section of tubing becomes "egged" and a substantial amount of projected absorber area is lost.
- (5) A flat spiral coil design for an absorber surface has several deficiencies.

  Therefore, further development work on XLPE absorbers should concentrate on the more conventional parallel-tube geometries, and should evaluate the problems of headering the parallel tubes such as the dangers of stress concentrations and leaks that are inherent with headers.

(6) The absorptivity to solar radiation of a flat spiral coil of 0.3" diameter carbon-black-reenforced crosslinked polyethylene is about as absorptive to solar radiation as are typical black paint coatings.

#### RECOMMENDATIONS

- (1) Crosslinked polyethylene absorber surfaces in the form of conventional parallel tube designs should be developed as outlined in the response which Polyset, Inc. made to ERDA's PRDA EG 77 D 29 0003 submitted on April 26, 1977 to ERDA's Albuquerque Operations Office.
- (2) Service tests on both the flat spiral coil collector and on collectors having parallel-tubes-with headers, configurations should be performed to determine whether, or rather to what extent, there are practical difficulties associated with freeze-tolerant solar collectors made from crosslinked polyethylene that could not be anticipated by the freeze-thaw tests on the 10 ft. specimens.

Theoretical Performance of a Solar Collector having;

- Done glazing with 8% reflectivity, 17% absorbtivity, 75% transmissivity BTu/hr and 0.937 emissivity
- 2)31/2" of insulation with k = 0:0225 BTu/hr
- 3) an absorber surface with d= E= 0.97
- 4) spacing between glazing + absorber = 1/2"
- 5) 45° slope
- 6) 4"x4' glazing

In all cases assume 80°F ambient & no wind

Case 1 Absorber temp, = 80°F (ie"y" intercept conditions on performance curve)

Heat transfer coeficients from glazing at an assumed temperature of 90°F

he inside = 0.156  $\frac{(\Delta t)^{0.31}}{L^{0.07}} \times [1.0+0.001(t-50)] = 0.156 \times \frac{10^{0.31}}{0.5001} [1.0+0.001(90-50)] = 0.349 \frac{BTWhr}{0Fft}$ he outside = 0.33  $\frac{(\Delta t)^{0.25}}{L} = 0.33 \left(\frac{10}{4}\right)^{0.25} = 0.414 \frac{BTW/hr}{0Fft}$ hr in both directions = 0.173×  $E\left(\frac{T_1}{100}\right)^4 - \frac{T_2}{1000}\right)^4 = 0.173 \times 0.937 \left(\frac{550}{100}\right)^4 - \frac{540}{1000}\right)^4 = 1.05 \frac{BTW/hr}{0Fft}$ 

striking the glazing Basis: 100 BTU reflected absorbed in the glazing transmitted absorbed on first bounce in the absorber 75 ×0.97 = 72.75 75 × 0.03 = 2.25 reflected back to the glazing absorbed on first bounce in the alazing 2.25 × 0.17= 0.47 reflected back to the absorber 2.25 x 0.08 = 0.22 absorbed on second bounce in the absorber 0.22 × 0.97 = 0.21 (17+0.47) = 0.249+1.05 = 8,50 of heat transferred from glazing to absorber of light energy absorbed by absorber 72.75+0.21 =7296

72.96+8.50 =81.46 BTU absorbed by absorber both directly + indirectly

Case 2 Stagnation Conditions le 2" intercept conditions on performance cum Assume: Plate temp. = 270°F, Glazing temp. = 140°F hr plate = 0.173x, 97 ( 450+270)4 - (450+140)4] = 0.173x, 97 [2839, 92-12.95] = 1,99 BTU/hr hr cover = 0.173x.937 [(460+140)4 - (460+140)4] = 0.173x,97[1296-850.3] = 1.20 BTU/hr hcp-c = 0.156 (1+0.001(+50)) = 0.90 BTW/hr UT hope + hope + hoca + hoca hcc-A = 0.33 (At )= 0.33 (270-140)0.25 0.78 BTU/A  $\frac{1}{U_{T}} = \frac{1}{1.99 + 0.90} + \frac{1}{1.2 + 0.78}$ Back Loss Coefficient = UB = 0.0225 = 0.0077 /UT = 0.346 + 0.505 = 0.851 UT = 1.174 BTUAT Total Loss Coeff. = 1.174 + 0077= 1.18 BTU/hr Calculate Solar Flux needed to give a 270°F Stagnation temp 1.18 x 1 x (270-80) = 224 BTU absorbed = I x 0.8146 x .97 I = 283.7 BTU/hr AT = 270-90 = 0.67 = "x" intercept on Performance Curve

```
<u>Case 3</u> Establish an Intermediate Point between the "y"Intercept and the "z" Intercept on the Performance Curry
   Assume: Plate at 160°F Cover at 115°F 280 BTM 2 Solar flux
 hr plate = 0.173 x 0.97 (460+160) - (460+115)47 = 0.173 x 0.97 (1477.6-1043.1) = 1.43 BTU/hr
 hr cover = 0.173 x 0.937 \[ \left(\frac{440+85}{100}\right)^4 - \left(\frac{440+85}{100}\right)^4 \right] = 0.173 \times 0.437 \left(1003.1-850.3) = 1.12 \frac{\text{BTU/hr}}{05 \text{C+2}}
 h_{c,p-c} = 0.156 \frac{(\Delta +)^{0.31}}{20.07} \left[ 1.0 + .001 (+-50) \right] = 0.156 \frac{45^{0.31}}{0.5007} \left[ 1.0 + .001 (10-50) \right] = 0.59 \frac{BTU/hr}{0.507}
hc, c-A = 0.33 (A+)0.25 = 0.33 x (115-80)0.25 = 0.57
\frac{1}{U_T} = \frac{1}{h_{r,p-c} + h_{c,p-c}} + \frac{1}{h_{r,c-A} + h_{r,c-A}} = \frac{1}{1.43 + 0.59} + \frac{1}{1.12 + 0.57} = 0.495 + 0.592 = 1.087
 UT = 0.9202 Back Loss Coefficient = 0.0077
                     Total " = 0.93 BTU/hr
  Heat Losses = 0.93 x (160-80) = 74.2 BTU
  Solar Flux = 280 BTU
  Heat Absorbed = 280 x. 8146 x. 97 = 221.2 BTU hr ft2
 \Delta T = \frac{160-80}{290} = 0.29
```

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