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**VEGETABLE OILS: LIQUID COOLANTS FOR SOLAR HEATING AND
COOLING APPLICATIONS**

Semi-Annual Report for September 29, 1978—March 31, 1979

By
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March 1979

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U.S. Department of Energy

Solar Energy

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I. ABSTRACT

As high temperature solar energy systems are developed for applications such as refrigeration, heating, cooking, electrical power generation, and industrial processes, the need for inexpensive, reliable heat transfer media (liquid coolants) will become more intense. At present petroleum distillates and synthetic coolants are being utilized for these purposes. Since the use of these substances represents a drain on our natural fuel resources, it has been proposed that vegetable oils, renewable byproducts of agriculture processes, be investigated for possible use as liquid coolants. This research project was implemented for the purpose of researching such a use.

During the conceptual phase of the study, it was recognized that very little data are readily available for predicting the results of using vegetable oils as liquid coolants in high temperature solar systems. Therefore, the major thrust of the project will be to investigate several thermophysical properties of the four vegetable oils selected. Vapor pressures, specific heat, viscosity, density and thermal conductivity will be determined over a range of temperatures for corn, soybean, peanut and cottonseed oil. ASTM standard methods will be used for these determinations.

Boiling point data and viscosity data have been determined for each of the four oils selected. Chemical analyses have been performed on samples of each oil. The samples were collected before and after each boiling point experiment so that any changes in composition could be noted. The tests include iodine number, fatty acid, and moisture content determination.

Future research will include density measurements, specific heat and thermal conductivity determinations. Stability and corrosion tests will also be performed in order to investigate long term usage of these oils in conventional solar systems.

II. NARRATIVE

A. Research Objectives

The research objectives include several tasks required to define the use of vegetable oils in high temperature (greater than 180°F, 82.2°C) solar systems:

1. Perform literature survey for existing thermophysical property data;
2. Perform thermophysical property experiments;
3. Perform stability and corrosion experiments;
4. Establish potential for each oil as a liquid coolant for solar applications;
5. Evaluate cost effectiveness of the selected oils;
6. Analyze pumping power required to transfer a unit of heat energy with the selected vegetable oils.

The thermophysical properties are to be determined at a pressure of about one atmosphere at 20°F (11.1°C) increments from 70°F (21.1°C) up to the boiling point. Cottonseed, corn, peanut, and soybean oil were selected as the tentative candidates for use in solar systems.

B. Literature Survey/Formation of Research Plan

Previous research was conducted to determine the thermal properties of many of the commercially available natural oils. Published literature indicates that these data are not readily available in the temperature range of 70°F (21°C) to 700°F (371°C). Preliminary studies and existing data did indicate that fluid properties, such as viscosity, freezing point and density are in an acceptable range to indicate some potential in solar applications. A study by Wakeman and Magne (1,2) defined flow properties of cottonseed oil between 86°F (30°C) and 410°F (210°C). Their data indicated that the viscosity of this oil fell in the range of viscosities given for commercially acceptable liquid coolants. Data on flow properties above 410°F (210°C) had not been adequately developed.

Vegetable oils have high temperature flammability properties. Jameson (3) has tabulated smoke, flash and fire points for several of these oils. Flash points have been recorded at approximately 620°F (327°C) and fire points at 670-680°F (354-360°C). The spontaneous ignition point is also an important property, and published data show that some of these oils (e.g. soybean) have ignition points as high as 833°F (445°C).

It is believed that the vegetable oils are much safer than other fluids. These oils are commonly used in or around food products and are non-toxic. Although some of these oils will produce corrosion in copper pipes if oxygen is present, there is no evidence to date that these oils will accelerate corrosion in closed systems, provided moisture content is low.

Their relative low cost is another attractive feature for using vegetable oils in solar systems. Prices range from \$3 to \$5 per gallon while petroleum based coolants and synthetic oils as much as \$30 per gallon.

Research at the University of Florida (4,5) has, in part, validated the use of these vegetable oils as liquid coolants. The use has dealt mainly with air conditioning and heating. In each case the research was designed to prove the feasibility of the concept and not to provide long term testing of the liquid coolant. If these oils are to realize widespread use in solar systems then reliable design data must be made available. This project has been designed to provide these data.

The more common oils; peanut, soybean, corn, and cottonseed, were selected for this study. A series of experiments were proposed in order to measure the thermophysical properties necessary for the proper design of solar equipment utilizing these oils. Coincident with these experiments, stability and corrosion tests will be run. The purpose of these experiments is to extend the thermophysical property data to higher temperatures and obtain the supplemental data needed to perform engineering calculations related to heat transfer and pumping power.

C. Experimental Facilities

The experiments performed under this contract were implemented at the University of Florida's Solar Energy and Energy Conversion Laboratories. As these experiments involved several physical chemistry procedures, the selective coating laboratory facilities provided an excellent base for this research. The laboratory provided air, gas, water, special ventilation requirements, sewage, and electrical power requirements. The laboratory facilities can accommodate the most delicate analytical equipment and yet still provide space for bench tests, such as the corrosion and stability tests required of this research.

D. Experimental Equipment/Procedures

This study involved thermophysical property determinations for the oils at a pressure of approximately one atmosphere. Data were to be collected at 20°F increments from 70°F (21.1°C) to the boiling point.

1. Boiling Point

Using ASTM standard method D1120 -72, Standardized Method for Boiling Point of Engine Coolants(6), the boiling point was determined for each of the four oils selected for study; corn oil, peanut oil, cottonseed oil, and soybean oil. The apparatus used for these experiments is illustrated in Figures 1 and 2. Figure 1 is a schematic of the apparatus. The carbon dioxide (CO₂) gas was used to provide an inert atmosphere during boiling experiments. A more detailed discussion of the rationale for the use of CO₂ may be found in the Experimental Results section of this report.

Even though ASTM D1120-72 is specifically directed toward boiling points for automobile coolants (engine coolants that are miscible with water), the procedure lends itself easily to the determination of vegetable oil boiling points with the modifications shown in Figure 1. In the case of this research, a carbon dioxide atmosphere was used above the oil to prevent oxidation of the oil. The flask size was increased to 1000 milliliters to accomodate the CO₂ apparatus and the sample size was increased to 100 milliliters accordingly. The condenser was allowed to operate dry as there was sufficient heat transfer area to provide reflux with air as the coolant. Using water in the condenser subcooled the oil to the point that the reflux drops would create turbulence and fumes when they hit the hot boiling fluid in the flask.

Boiling Point Test Apparatus

Figure No. 1

Figure No. 2
Boiling Point Apparatus

The oils were heated at a rate sufficient to raise the temperature approximately 1°F every 10 seconds until boiling and reflux were realized. After reflux was established, the temperature and barometric pressure were recorded. The boiling temperature reading was corrected for thermometer inaccuracy and barometric pressure. The Sydney Young equation was used to correct for deviations in barometric pressure.

2. Viscosity

For the determination of the vegetable oil viscosity, three methods were considered, ASTM D2515-74, ASTM E102-62 (reapproved 1973), and ASTM D88-56. In light of the temperature range proposed for the study, the availability of equipment, and the expected viscosity range, method D88-56 was selected for the tests. This method is entitled "Standard Method of Test for Saybolt Viscosity (7)." A GCA/Precision Scientific Universal Saybolt viscometer (model 74965) was used for these tests (Figures 3,4). The viscometer tube and universal orifice are shown in detail in Figures 5 & 6. The timer was a Technilab Instruments Rapid Reset Timer, accurate to 0.1 second. The timer was calibrated against a standard timepiece. The Universal Saybolt viscosity in seconds and the corresponding oil temperature in °F was recorded for each oil. The temperature readings were corrected for thermometer inaccuracies.

3. Other Determinations

At the time of this report preparation, the viscosity measurements had just begun. Following these viscosity tests, the methods listed below will be implemented in order to obtain other thermophysical properties of the vegetable oils:

Figure No. 3

Figure No. 4
Universal Saybolt Viscometer

Sectional View of SAYBOLT Viscometer
Figure No. 5

Figure 6

Universal Saybolt Viscometer Tube and Orifice

Specific heat (8)	D2766-71
Density (9)	ANSI/ASTM D1481-62
Thermal conductivity (10)	D2717

Vapor pressure data and density data will be used to extrapolate the thermophysical properties to higher pressures. The vegetable oils will be subjected to cycling from 300°F to some higher temperature in a closed system to study the influence of thermal cycling on the properties of the oil. Corrosion tests will also be performed during this time period.

4. Chemical Analyses

Samples of oil were taken before and after each property determination and subjected to chemical analyses. Each sample was tested for iodine number, fatty acid content, and moisture content. These data provided information related to the possible degradation of the sample during the property determinations. At the high temperatures encountered during these property experiments, it is possible that the chemical bonds of the hydrocarbons comprising these oils could break or react with other substances such as oxygen or water.

The chemical analyses were performed by an independent research laboratory. The following methods were utilized:

Iodine Absorption Number (18)	Methods 28.018 Hanus Method
Free Fatty Acid Content (18)	Method 28.029
Moisture and Volatile Matter (18)	Method 28.002

E. Experimental Results

Table 1 presents the boiling point data obtained by the procedures outlined in the previous section. These data have been corrected for thermometer inaccuracies and for barometric pressure.

Table 2 presents the viscosity data for the four vegetable oils in centipoise and Universal Saybolt seconds. These data were obtained using the procedures discussed above. ASTM D2161 was used to make the conversion from Saybolt seconds to centistoke. In order to calculate the viscosities in centipoise an empirical relationship (11) was used to determine the densities of soybean, cottonseed and peanut oil. As there was no density data for corn oil, the conversion to centipoise was not made. When the density data for these oils has been determined experimentally, the centipoise values will be recalculated.

Table 3 illustrates the results of the chemical analyses performed on the four oils. These data include the moisture content, fatty acid content, and the iodine number for each of the four oils tested.

TABLE 1
BOILING POINT DATA*
VEGETABLE OILS

	Mean Boiling Point (Uncorrected) $t_{bp}, ^\circ F$	Barometric Pressure, mmHg	Mean Boiling Point (Corrected) $t_{bp}, ^\circ F$	Standard
Soybean Oil	708.6 (375.9°C)	771.5	707.3 (375.2°C)	2.6
Cottonseed Oil	712.8 (378.2°C)	772.6	711.4 (377.4°C)	0.9
Peanut Oil	719.4 (381.9°C)	772.6	718.0 (381.1°C)	0.8
Corn Oil	707.8 (375.4°C)	770.7	706.6 (374.8°C)	0.2

* ANSI/ASTM D 1120-72

TABLE 2

VISCOSITY DATA

VEGETABLE OILS

	Oil Temperature, °F	Universal Saybolt Viscosity, Seconds	Centipoise
Soybean Oil	85.8	236	46.62
	103.9	190	37.22
	125.8	153	29.66
	151.0	134	25.66
	174.5	121	22.92
	200.0	112	20.95
	224.5	107	19.78
	249.5	102	18.62
Cottonseed Oil	85.7	258	50.33
	104.2	196	37.91
	126.0	155	29.69
	151.0	135	25.56
	174.9	121	22.66
	200.0	112	20.73
	224.5	106	19.38
	249.2	101	18.25
Peanut Oil	85.7	247	48.52
	103.4	195	37.99
	125.7	160	30.86
	150.0	134	25.54
	172.5	122	23.01
	198.0	120	22.36
	222.0	107	19.70
	246.5	101	18.38
Corn Oil	86.7	243	
	103.8	194	
	125.2	158	
	150.0	140	
	174.0	125	
	199.5	112	
	224.0	105	
	249.5	101	

TABLE 3
CHEMICAL ANALYSIS
OF VEGETABLE OIL
SAMPLES

		%Moisture	%Free Fatty Acid	Iodine Number
Soybean Oil	Control	0.32	0.50	127
	Exp. No.5	2.80	33.00	75
	Exp. No.7	0.81	13.7	85
Cottonseed Oil	Control	0.07	0.28	81
	Exp. No.12	2.30	5.40	62
Peanut Oil	Control	0.05	0.28	85
	Exp. No.19	3.60	9.30	67
Corn Oil	Control	0.01	0.28	108
	Exp. No.24	0.39	2.8	82

F. Discussion of Results/Statistical Analysis

The boiling points reported in Table 1 were taken at approximately 760 mm Hg pressure under an inert atmosphere of carbon dioxide. Without this carbon dioxide, the vegetable oils reacted with atmospheric air, resulting in a color change of the liquid and spontaneous ignition of the vapors. A report prepared for this study by Gerald Doddington (11) provides information on the chemical degradation which may result from excessive heating and oxidation of vegetable oils.

Several possible chemical reactions may take place, including oxidation, isomerization, polymerization, and pyrolysis. These reactions can occur spontaneously with the addition of heat; subsequent changes in chemical composition can greatly affect the heat transfer properties of the vegetable oils as well as the properties related to pumping power.

Oxygen reacts more readily with unsaturated organic acids than with saturated acids. Since the oils are 70-80% unsaturated acids, this is certainly a potential problem. At low temperatures, oxidation will occur at a relatively slow rate. This rate of oxidation will remain fairly low for some period of time, called the induction period. After the induction period, oxidation rates will increase greatly. The length of the induction period is halved for every 18°F (10°C) increase in temperature.

There are two methods of reducing oxidation: 1) heat the oil in a closed system or under an inert atmosphere and 2) use natural antioxidants such as lecithin or gossypol (12). Typically a very small quantity of gossypol will have dramatic effects on reducing the rate of oxidation.

In future studies the effects of these antioxidants on the thermophysical properties of the vegetable oils and their effectiveness in preventing oxidation will be investigated.

Generally, oxidation reactions would be expected to break the oils into lower molecular weight substances; however, oxidation is also accompanied by polymerization which has the opposite effect. The overall effect is reported to result in an increase in viscosity (13). Polymerization can occur spontaneously within vegetable oils with the addition of sufficient heat. In the temperature range of 482 to 572°F (250 to 300°C) the rate of polymerization increases rapidly. This thermal polymerization can result in the formation of a variety of organic molecules, including ring types. Ralston (14), Anderson and Porter (15) elaborate on several of these polymerization reactions.

Isomerization is another reaction associated with heating. This reaction results in a rearrangement of atoms within an organic molecule so that the chemical formula does not change. This may involve a change as simple as moving the location of a double bond. Eckey (16) cites experimental evidence that heating oleic acid to 356°F (180°C) will result in approximately 8% isomerization; thereby changing the thermophysical properties of the oil.

The term generally used to describe the decomposition of oils due to the addition of heat is pyrolysis. In some vegetable oils, pyrolysis occurs at temperatures as low as 572°F (300°C). At temperatures above 752°F (400°C), this decomposition is extensive, resulting in the production of compounds such as ketones, water, and miscellaneous gaseous and liquid hydrocarbons. Egloff and Morrell (17) obtained a 54% yield of gasoline through pyrolysis of cottonseed oil.

In order to determine the extent of degradation, if any, which occurred during the boiling point determinations, samples of the vegetable oils were taken before and after each test. The samples were then subjected to chemical analysis. The results of these tests were reported in Table 3. Typical ranges of iodine values for the oils studied are given in Table 4.

TABLE 4
TYPICAL IODINE VALUES
FOR SELECTED OILS

<u>Vegetable Oils</u>	<u>Iodine Values</u>
Soybean	117-141
Cottonseed	99-113
Cottonseed (hydrogenated)	78
Peanut	84-102
Peanut (hydrogenated)	63
Corn Oil	103-128

A comparison of the data in Table 4 with that in Table 3 for the control samples indicates that the iodine numbers for all but one of the oils falls within the expected range. Only cottonseed oil is low and out of the range. When this value is compared with that reported for hydrogenated cottonseed oil, it may be summarized that this oil sample was slightly hydrogenated.

After the oil was subjected to a boiling point determination, a sample was taken and chemical analyses were performed. These data are also reproduced in Table 3. The data for experiment No. 5, soybean oil, are for a boiling point determination in an environment of air; a color change was observed when the oil was heated. The data for experiment 7 were for the determination in an environment of CO_2 . The iodine number in each case is lower than the control value, with the sample from experiment 5 exhibiting the lowest value. This is an indication that oxidation may have occurred. The fact that the sample for experiment No. 7 is also lower than the control value illustrates that other chemical reactions such as pyrolysis may have also taken place. In each of the other experiments reported for the oils, the iodine value of the sample taken after the boiling point determination (in an inert atmosphere) was lower than the control.

The values given for percent moisture and percent free fatty acids are also indicative of trends observed when oils degrade due to overheating. During the boiling point determination, free fatty acids and moisture were produced, which indicated that the oils were breaking down. Tests will be performed during the next phase of this research in order to define the specific effects of increasing temperature on the thermophysical properties of the oils.

The viscosity data given in Table 2 are for temperatures ranging from approximately 85°F (29.4°C) to 250°F (121°C). These data were collected using a conventional viscometer. As soon as these data are verified, additional data will be gathered for temperatures up to the smoke point (Table 5). At this temperature, modifications will be implemented so that the viscometer's range may be extended to the boiling point.

TABLE 5
SMOKE, FLASH AND FIRE POINTS
SELECTED VEGETABLE OILS (14,19,20)

Oil	Smoke Point	Flash Point	Fire Point
Corn	352 ·F (178·C)	562·F (294·C)	655·F (346·C)
Cottonseed	420-450·F (216-232·C)	615-625·F (324-329·C)	670-685·F (354-363·C)
Peanut	450·F (232·C)	625·F (329·C)	685·F (363·C)
Soybean	410·F (210·C)	603·F (317·C)	670·F (354·C)

A standard error was calculated for each of the data points reported in Tables 1 and 2. There was no apparent reason to throw out the one data point which resulted in the 2.6 standard error for the soybean oil boiling point (Table 1).

G. Future Work and Research Goals

As discussed in earlier sections, the viscosity data must be extended to higher temperatures. The work required to accomplish this is already underway. Modifications to the Saybolt viscometer will be made as necessary to extend these data to the boiling point.

Construction of a temperature bath (fluidized bed type) is underway for use in the density measurements. Specific heat and thermal conductivity data will also be gathered for temperatures ranging from 70°F (21.1°C) to 700°F (371.1°C) for each of the subject oils.

Stability and corrosion tests will be performed to determine the effects of thermal cycling on the oil properties related to heat transfer and pumping power. Corrosion tests will be made to study the effects of these hot oils on various metals such as steel, copper and aluminum. The effects of these metals as catalysts in accelerating the degradation of the vegetable oils will also be investigated.

The four oils studied in this research will be compared to ethylene glycol solutions and water as heat transfer media. These comparisons will be made in the light of heat transfer characteristics, fluid flow properties, and cost.

III. Activities to Disseminate the Project Results

This report was distributed to the following list of people:

Donald J. Garcia, Albuquerque, New Mexico
D. K. Nowlin, Director, Albuquerque, New Mexico
Dr. Stephen L. Sargent, Washington, D.C.
Dr. Charles Bankston, Los Alamos, New Mexico
Solar Support Group, McLean, Virginia
G. L. Zachariah, Gainesville, Florida

Besides the normal dissemination of report copies, a report has been completed by a graduate student as partial fulfillment of the requirements for a Masters in Engineering. This report is listed as reference 11 in section V. A master's thesis on this work is also underway; several papers should be published reporting the efforts of this work. Within the next month, a newspaper story should be released on the vegetable oil research.

IV. REFERENCES

1. Magne, F. C. and Wakeman, H., Industrial and Engineering Chemistry, vol. 36, no. 6, pp. 568-570, 1944.
2. Magne, F. C. and Wakeman, H., Oil and Soap, vol. 21, pp. 347-49, 1944.
3. Jamieson, G. S., Vegetable Fats, and Oils, 2nd Ed., 1943.
4. Chari, R. K., "An Investigation of the Use of Solar Energy for Absorption Refrigeration", an unpublished master's thesis, University of Florida, 1958.
5. Elder, K. M., "A Solar System With Twenty Four Hour Food Preparation Capability," an unpublished master's thesis, University of Florida, 1975.
6. ANSI/ASTM D 1120-72, "Standard Test Method for Boiling Point of Engine Coolants", Published June 1972.
7. ASTM D88-56 (Reapproved 1968); "Standard Method of Test for Saybolt Viscosity", 1973 Annual Book of ASTM Standards, Part 17 pp. 21-27, Nov. 1973.
8. ANSI/ASTM D2766-71 (Reapproved 1976), "Standard Test Method for Specific Heat of Liquids and Solids", 1977 Annual Book of ASTM Standards, Part 24, pp. 643-651, 1977.
9. ANSI/ASTM D1481-62, "Standard Test Method for Density and Relative Density (specific gravity) of Viscous Materials by Lipkin Bicapillary Pycnometer", 1978 Annual Book of ASTM Standards, Part 40, pp 435-441, 1978.
10. ASTM D2717, "Standard Test Method for Thermal Conductivity of Liquids", 1977 Annual Book of ASTM Standards, Part 24, pp. 638-641, 1977.
11. Doddington, G. E., "Forced Convection Heat Transfer Coefficients and Pumping Power Requirements for Organic Oils Flowing Through Smooth Tubes," a report prepared as part of the requirements for a Master of Engineering, University of Florida, December 1978.
12. Royce, H. D.; and Lindsey, F. A. Jr., Industrial and Engineering Chemistry, vol. 25, p. 1047, 1933.
13. Bailey, A. E., Industrial Oil and Fat Products, Interscience Publications, 1945.
14. Ralston, A. W., Fatty Acids and Their Derivatives, John Wiley and Sons, Inc., 1948.
15. Anderson, L. V. and Porter, J. V., Industrial and Engineering Chemistry, vol. 41, p. 741, 1949.

16. Eckey, E. W., Vegetable Fats and Oils, Reinhold Publishers, City, 1954.
17. Egloff and Morrell, Industrial and Engineering Chemistry, vol. 24, p. 1426, 1932.
18. Horwitz, Williams, Editor, Official Methods of Analysis of the Association of Official Analytical Chemists, Published by the Association of Official Analytical Chemists, Washington, D.C., 12th Ed., 1975.
19. Adams, H. E., and Powers, P. O., Industrial and Engineering Chemistry, vol. 36, p. 1124 1944.
20. McGraw-Hill Book Co., International Critical Tables, 1926.