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THE USE OF ORGANIC WORKING
FLUIDS IN RANKINE ENGINES

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THE USE OF ORGANIC WORKING FLUIDS IN RANKINE ENGINES

INTRODUCTION

The original working fluid for Rankine cycle engines was water, and this is still used in power plants and other applications of high temperature Rankine engines. Water is plentiful, inexpensive, and can provide better cycle efficiencies than any other fluid. However, the low molecular weight of water requires the use of multi-stage expanders to obtain high cycle efficiency. For Rankine engines with maximum temperatures below 400°C, fluids with higher molecular weights than water can provide high cycle efficiency in less complex and less costly single-stage expanders. In this context, the word "fluid" has the connotation of a substance which undergoes liquid to vapor and vapor to liquid phase transformations in order to effect a Rankine cycle between specified temperature limits. In principle, practically any thermally stable substance could be used in a Rankine cycle, within the limitations imposed by the containment materials and operating conditions. For example, substances which are solids at environmental temperature and pressure have been used in high temperature cycles, and substances which are gases at environmental temperature and pressure have been used in very low temperature cycles.

This paper is primarily a compilation of state-of-the-art data on the use of organic working fluids in operational Rankine engines, and was prepared in connection with U.S.

Department of Energy Contract No. DE-AC03-79CS-30202. The work was directed in particular to determination of the maximum temperatures used for various working fluids in operational Rankine engines, and identification of thermal instability and chemical reaction problems related to these temperatures. The data were obtained from the literature and by correspondence with manufacturers and researchers in the U.S. and other countries.

ORGANIC WORKING FLUIDS

The use of organic working fluids in Rankine engines is not a recent concept. The use of diphenyl oxide, for example, was reported in 1926 [1]. Over the following half-century, many organic fluids and water-organic mixtures have been identified, proposed, and used in Rankine engines for various terrestrial and space applications [2-35]. Some of these fluids have been subjected to extensive dynamic testing for thermal stability [7, 9, 10, 11, 13, 23] (mono-isopropyl biphenyl, biphenyl, Dowtherm A, CP-9, R-22, CP-34, pyridine/water azeotrope) in test loops designed for this purpose. Others have been subjected to static tests in various types of calorimeters, capsules, and sealed tubes [13, 14, 36]. An important result of capsule testing has been an indication of a strong dependence of the fluid degradation rate on the presence of oxygen [13]. In order to eliminate oxygen both the fluid and the system materials must be degassed prior to charging with the fluid and the system must be sealed against in-leakage of air.

The most widespread use of organic fluids in thermodynamic cycles occurs in their use as refrigerants in refrigeration and air conditioning equipment. The vapor

compression cycle used in such equipment is a reversed Rankine engine cycle. An extensive literature exists on the thermodynamic and physical properties of many organic refrigerants, refrigerant system chemistry, moisture and contaminants, and lubricants, as exemplified by the ASHRAE Handbooks [56, 57].

Refrigeration systems and Rankine engines can fail as a result of the chemical decomposition of the working fluid. In addition to making replacement of the working fluid necessary, chemical decomposition of the fluid can produce non-condensable gases which degrade the heat transfer rate in the condenser, as well as liquid and gaseous compounds which have a corrosive effect on the system materials. The decomposition may be caused by the reaction of the organic fluid and the lubricating oil in the presence of metals and contaminants, or it may be caused by pyrolytic instability of the organic fluid or the oil, or some combination of these.

Chemical reactions are temperature dependent and usually increase by a factor of two or more for every 10°C increase in temperature [56]. Under ordinary operating conditions in vapor compression refrigeration equipment, chemical reactions involving the refrigerants proceed at negligible rates except at hot spots such as compressor discharge valves. However, in Rankine engines and high temperature heat pumps [58], wherein the maximum operating temperatures and hot spot temperatures are much higher than in refrigeration equipment, the potential for chemical reactions to occur at unacceptably high rates is much greater.

LUBRICATING OIL EFFECTS

With the exception of a few oil-free engines [31], organic fluids must co-exist with lubricating oil in Rankine engines. The selection of a suitable oil requires careful consideration of the desired physical and chemical properties, as well as the working fluid and materials of construction to be used. Numerous investigations of the behavior of oils in contact with organic fluids have been conducted [59, 60, 61, 62, 63, 64]. In general, these studies have shown that some oils are more stable toward organic fluids than others, with increased temperature accelerating the refrigerant-oil reaction. The reaction rate is also dependent on the kinds of metal in contact with the oil and organic fluid, the amount of air and moisture present, and the additives present in the oil [65].

When in contact, an organic fluid and a lubricating oil have a property known as mutual solubility. Organic fluids may be classified as completely miscible, partially miscible, or immiscible according to their mutual solubility relations with lubricating oils. In the case of partial miscibility, the mutual solubility is a function of the fluid/oil ratio and increases with increasing temperature. Above a certain temperature, the critical solution temperature, the fluid and oil are completely miscible.

The dissolving of an oil in an organic working fluid affects the thermodynamic properties of the fluid. The vapor pressure of an organic fluid-oil solution at a given temperature is always less than the vapor pressure of the pure organic fluid at that temperature. The magnitude of the vapor pressure depression depends on the fluid/oil ratio, being quite small when the fluid/oil ratio is large, and vice versa.

Similarly, the dissolving of an organic working fluid in an oil affects the properties of the oil. In particular, the viscosity of an organic fluid-oil solution at a given temperature is generally less than the viscosity of the pure oil at the same temperature.

SELECTION OF WORKING FLUID

The choice of an organic working fluid for a Rankine engine is governed primarily by the physical and thermodynamic properties of the fluid. A candidate fluid should be chemically stable at the maximum expected cycle temperatures and in contact with the engine materials, lubricating oil, and unavoidable contaminants. Preferably, it should also be non-corrosive, non-flammable, non-toxic, and low in cost.

Although much of the information developed in the refrigeration industry on the use of organic fluids in thermodynamic cycle equipment is directly applicable to Rankine engine design, additional information specific to the design must be developed. Because of the higher temperatures, the thermal stability may be a more important factor in the selection of a working fluid than the thermodynamic properties. Extensive tests in capsules or sealed glass tubes are commonly performed in order to determine the chemical and thermal stability of candidate fluids in the vicinity of the maximum expected engine temperature, and in contact with the engine materials and probable contaminants. Less common are long-duration dynamic tests in which a candidate fluid is circulated in a closed loop to simulate engine temperature, pressure, and material conditions.

Dynamic testing is highly desirable prior to selection of a final design and construction of an engine. Although dynamic testing is expensive and will not necessarily result in the elimination of all flaws in the final design, it provides a lower risk approach than simply designing and constructing an engine for a particular working fluid and testing it.

A partial list of organic fluids which have been used or proposed for use in operating Rankine engines is given in Table 1. The table shows the molecular weight, critical point, freezing point at atmospheric pressure, the approximate thermal stability limit, the vapor pressure at 40°C or the saturation temperature at 10 kPa, and whether the fluid is "wetting" or "drying." "Wetting" fluids are those which become saturated or partially condensed upon expansion; "drying" fluids are those which become superheated upon expansion. This property is related to the number of atoms per molecule of the fluid [4]. The saturation temperature at 10 kPa is an approximate lower limit for condensing with surface condensers. For pressures below 10 kPa, jet condensers are preferable to surface condensers.

The thermal stability limits given in Table 1 are, as indicated, approximate. The results of tests of thermal stability for a given fluid, as reported in the literature, show considerable variation, depending on the test equipment, the procedure, the test duration, and the materials-fluid contact. The thermal stability limit of an organic fluid cannot be defined in a precise manner which would be applicable to all possible uses. Test results generally indicate a chemical decomposition rate as a function of temperature and the test conditions. Thus, the thermal stability limit values in Table 1 are representative of temperatures at

which the chemical decomposition rate would be unacceptably high for use in Rankine engines.

OPERATIONAL EXPERIENCE

Table 2 provides data on about 2,150 operational Rankine engines which use sixteen different organic working fluids. Not all such engines are included, but the list is representative of the range of sizes, fluids, and applications. Many of these are (or were) experimental engines with only a small running duration. The power output covers a range of 0.1 kW to 1120 kW, and the expanders are predominantly turbines. Information on the total operating time was not available for all of the engines listed. In general, these are values as of September 1979 or some earlier date, depending on whether the information was obtained from a private communication or from the literature.

Some twenty different engine manufacturers are represented in Table 2. In terms of number of engines and operating time, the major manufacturer of organic Rankine engines is Ormat Turbines of Yavne, Israel, with 2,000 engines and 18,000,000 operating hours. The working fluid is trichlorobenzene.

The published literature and the private communications report few operating problems directly related to the fluid itself. This may be due either to the fluids having a high degree of thermal and chemical stability for the operating conditions or to the engines not having been operated long enough for fluid-related problems to become evident. The maximum fluid temperatures given in the table are the reported boiler temperatures. Exposure of fluids to higher

temperatures which may occur at hot spots is rarely mentioned in the literature. The listed fluids for which some fluid decomposition and/or corrosion occurred were R-11, R-113, trichlorobenzene, CP-25 (toluene), and alcohol/water.

RECOMMENDED ORGANIC FLUIDS R&D

The Rankine engine manufacturers listed in Table 2 generally have on-going efforts in organic working fluid research and development covering thermal and chemical stability, corrosion mechanisms, compatible materials and lubricants, additives, fluid cost, thermodynamic and thermo-physical properties, toxicity, and flammability. The extent of these efforts varies from limited investigation of one fluid to extensive investigations of many fluids. Much of this work is privately funded so that the results are largely proprietary.

Correspondence with representatives of a number of manufacturers indicated that further research is needed for presently known fluids, although the degree to which this is required for a particular fluid depends on the amount of reliable data already available on the fluid. Some recommended that efforts be made to identify or synthesize new fluids with thermodynamic, thermo-physical, and other properties which are more compatible for Rankine engine use than those of presently used fluids. The temperature range of interest is 100 to 400°C. Recommendations were also made for research and development on such fluid-related problems as more effective filters and dryers and corrosion inhibitors.

TABLE 1. RANKINE CYCLE ORGANIC WORKING FLUIDS

FLUID	CHEMICAL FORMULA*	MOLECULAR WEIGHT	CRITICAL PT. C/kPa	FREEZING PT. C	APPROX. THERMAL STABILITY LIMIT C	VAPOR PRESS. AT 40°C, kPa	SAT. TEMP. AT 10 kPa, C	WETTING (W) DRYING (D) NEITHER (N)
R-11	CCl_3F	137	198/4413	-111	120	159		N
R-21	CHCl_2F	103	178/5102	-135		276		W
R-22	CHClF_2	86	96/4978	-160	200	1455		W
R-113	$\text{C}_2\text{Cl}_3\text{F}_3$	187	214/3441	- 35	175	76		D
R-114	$\text{C}_2\text{Cl}_2\text{F}_4$	171	146/3261	- 94	175	317		D
R 133a	$\text{C}_2\text{H}_2\text{ClF}_3$	118	152/4068	-106	200	310		N
CP-9(a)	$\text{C}_{15}\text{H}_{16}$	196	522/2448	- 55	370		200	D
CP-25(b)	C_7H_8	92	321/4254	- 95	480	7		D
CP-27(c)	$\text{C}_6\text{H}_5\text{Cl}$	113	359/4523	- 55	320	7		D
CP-32(d)	$\text{C}_5\text{H}_5\text{N}$	79	347/5633	- 42	370	7		D
CP-32/ Water(e)	0.23 $\text{C}_5\text{H}_5\text{N}$	33	366/8964	- 18	400	14		D
	0.77 H_2O							
CP-34(f)	$\text{C}_4\text{H}_4\text{S}$	84	307/5461	- 40	290	21		<66C: W >66C: D

TABLE 1. (CONTINUED)

FLUID	CHEMICAL FORMULA*	MOLECULAR WEIGHT	CRITICAL PT. C/kPa	FREEZING PT. C	APPROX. THERMAL STABILITY LIMIT C	VAPOR PRESS. AT 40°C, kPa	SAT. TEMP. AT 10 kPa, C	WETTING (W) DRYING (D) NEITHER (N)
FC-75(g)	$C_8F_{16}O$	416	227/1607	- 62	320	7		D
FC-88(h)	C_5F_{12}	288	150/2131	-115	200	140		D
F-85(i)	0.85 CF_3CH_2OH 0.15 H_2O	88	240/6412		290	21		W
Dowtherm A(j)	0.265 $(C_6H_5)_2$ 0.735 $(C_6H_5)_2O$	166	499/3241	- 48	370		~140	D
Biphenyl	$(C_6H_5)_2$	154	498/3289	69	370		~140	D
Allied P-1 D	$C_{10}F_{22}O_2$	570	243/1186	- 85	370	2	~75	D

*Mixture fractions on mole basis

- (a) Monoisopropyl biphenyl $(C_6H_5)(C_6H_4)-CH(CH_3)_2$
- (b) Toluene
- (c) Monochlorobenzene
- (d) Pyridine
- (e) Azeotrope of pyridine and water
- (f) Thiophene
- (g) Perfluoro-2-butyltetrahydrofuran
- (h) Perfluoropentene
- (i) Fluorinol (Trifluoroethanol/water mixture)
- (j) Eutectic of biphenyl and phenyl ether

TABLE 2. OPERATIONAL RANKINE ENGINE DATA

Working Fluid	Engine Energy Source	Type	Expander		Manufacturer	Driven Equipment	No. of Operating Engines	Total Operating Time	Max. Fluid Temp. C	Reported Fluid Related Operating Problems	Location	Reference
			Power kW	Speed rpm								
R-11	Solar	Screw	5-100	1500-1800	SOFRETES	Water pumps, generators	70	>4,000 hrs.	95	None	Various countries	44,80
R-11	Solar	1-Stage impulse turbine	35	5,500	Thermo-Electron	Irrigation pump	1	>100 hrs.	86	None	Bakel, Senegal	45,73
R-11	Solar	1-Stage radial inflow turbine	1.7 12 37	60,000 30,000 18,000	AiResearch	Vapor compressor	4 2 2		95 95 95	None	Various	31,55
R-11	Solar	1-Stage radial inflow turbine	34	11,950	Barber-Nichols	Vapor compressor	1	2,000 hrs.	86	None	Los Alamos, NM	68,69,79
R-11	Solar (a)	1-Stage radial inflow turbine	16	40,400	Hamilton Standard (sub to UTRC)	Vapor compressor	1	25 hrs.	150	None	Hartford, CT	49
R-11	Electric (b)	Multivane positive displacement	2.3	1200-1800	General Electric	Vapor compressor, dynamometer (b)	None at present	1,200 hrs. (b)	105	Rapid decomposition of R-11 above 105°C with oil present	Evendale, OH Valley Forge, PA	71
R-11	Steam generated by Diesel exhaust	Radial inflow turbine	335	9,500	MTI	Generator	1		88	None	Rockville Center, NY	76,77
R-22		Reciprocating	3	3,600	Thermo-Electron	None	1	>100 hrs.	230	None	Waltham, MA	73
R-113	Solar	1-Stage radial inflow turbine	37	30,700	Barber-Nichols	Irrigation pump	1	500 hrs.	135	None	Gila Bend, AZ	40,79
R-113	Solar	1-Stage radial inflow turbine	19	36,300	Barber-Nichols	Irrigation pump, generator	1	600 hrs.	165	Corrosion due to moisture & air in-leakage, clogging of filters and valves	Willard, NM	38,39,72,
R-113	Solar	1-Stage radial inflow turbine	1.7 15	35,000 24,400	Barber-Nichols (sub to Honeywell)	Vapor compressor	2 7	 750 hrs.	80 80	None None	Various Texas, Georgia, Minnesota, Kansas	35,51,52,

TABLE 2. (Continued)

Working Fluid	Engine Energy Source	Expander		Power kW	Speed rpm	Manufacturer	Driven Equipment	No. of Operating Engines	Total Operating Time	Max. Fluid Temp. C	Reported Fluid Related Operating Problems	Location	References
		Type											
R-113	Solar	1-Stage radial inflow turbine	63.4	22,400	Barber-Nichols	Vapor compressor	2	100 hrs.	135	None		Minneapolis, MN	53,79
R-113	Solar	1-Stage radial inflow turbine	20	20,100	MTI (sub. to Carrier)	Vapor compressor	1		150	None		Syracuse, NY	32
R-113	Solar	Turbine	15	42,000	Dornier System	Generator	3	500 hrs.	110	Shaft sealing		Cairo, Egypt (2) Vienna, Austria(1)	50,66
R-113	Steam generated by Diesel exhaust	Turbine	60		Nigata Iron Works	Generator	1		70	None		Tokyo, Japan	41
R-114	R-114 vapor from process (c)	1-Stage impulse turbine	375	1,800	Worthington	Compressor	1	6 yrs.	120(d)	None		Portsmouth, OH	29
R-114	R-114 vapor from process (c)	1-Stage impulse turbine	375	1,800	Worthington	Compressor	1	10 yrs.	120(d)	None		Portsmouth, OH	29
R-114	R-114 vapor from process (c)	1-Stage reaction turbine	1050	4,800	Allis-Chalmers	Compressor	1	2.5 yrs.	120(d)	None		Oak Ridge, TN	29
R-114	R-114 vapor from process (c)	6-Stage impulse turbine	1120	1,200	TRW	Compressor	1	1.75 yrs.	120(d)	None		Oak Ridge, TN	29
R-114	Geothermal	Turbine	1000		Toshiba	Generator	1			None		Hokkaido, Japan	41
R-114	Solar, Oil	Helical Screw	20	7,500	Messerschmitt-Bolkow-Blohm	Generator	5	1000 hrs.	200	None		Germany, India, Algeria	43,67
CP-9	Gasoline	Impulse turbine	0.1	30,000	Aeronutronic	Generator	1		315	None		Newport Beach, CA	7
CP-25	Solar, Oil	Turbine	150	20,000	Messerschmitt-Bolkow-Blohm	Generator	1		450	Flammability		Munich, Germany	43

TABLE 2. (Continued)

Working Fluid	Engine Energy Source	Type	Expander			Driven Equipment	No. of Operating Engines	Total Operating Time	Max. Fluid Temp. C	Reported Fluid Related Operating Problems	Location	Reference
			Power kW	Speed rpm	Manufacturer							
CP-25	Solar, Gas	1-Stage impulse turbine	32	19,400	Sundstrand	Generator	1	2400 hrs.	300	Corrosion caused by moisture & air in-leakage, flammability, clogging of filters & valves	Sandia Laboratory, Albuquerque, NM	37,38,39
CP-25	Waste Gas	1-Stage impulse turbine	600	9,300	Sundstrand	Generator	5		240	None	Various	30
CP-25	Propane, Oil	1-Stage impulse turbine	1		Sundstrand	Generator	3	13,000 hrs.	385	None	Various	54
CP-34	Gas	Reciprocating	3	1,800	Thermo-Electron	Generator	1	>100 hrs.	290	None	Waltham, MA	13,73
FC-75	Oil	2-Stage turbine	50	6,500	Bertin et Cie	Generator	1	1,000 hrs.	250	None	Plaisir, France	70
FC-88	Solar, Electric (b)	2-Stage multivane positive displacement	2.3	1,625	General Electric	Vapor compressor, dynamometer (b)	1	1,000 hrs.	175	None	Valley Forge, PA	48,71
			7.9	1,625			2	(b)	175			
F-50	Diesel exhaust gas	1-Stage turbine	38	35,000	Thermo-Electron	Automotive	3	>100 hrs. each	315	None	Waltham, MA	73
F-50	Diesel exhaust gas	3-Stage turbine	38	60,000	Thermo-Electron	Automotive	1	>100 hrs.	315	None	Waltham, MA	73
F-85	Furnace exhaust	Turbine	670	11,100	Mitsui	Generator	1		290	None	Fukuyama, Japan	41
F-85		Reciprocating	3	1,800	Thermo-Electron	Generator	3	>100 hrs.	290	None	Waltham, MA	73
						Sweeper	1	each	290			
						Vehicle	2	290				
F-85		1-Stage turbine	3	70,000	Thermo-Electron	Generator Vehicle	1	>100 hrs. each	290	None	Waltham, MA	73
F-85		Reciprocating	112	1,800	Thermo-Electron	Automotive	1	>100 hrs.	315	None	Waltham, MA	73

TABLE 2. (Continued)

Working Fluid	Engine Energy Source	Expander				Driven Equipment	No. of Operating Engines	Total Operating Time	Max. Fluid Temp. C	Reported Fluid Related Operating Problems	Location	Reference
		Type	Power kW	Speed rpm	Manufacturer							
F-85		1-Stage turbine	450	18,000	Thermo-Electron	Generator	1	>100 hrs.	290	None	Waltham, MA	73
F-85		6-Stage turbine	450	12,500	Thermo-Electron	Generator	1	>100 hrs.	290	None	Waltham, MA	73
Trichlorobenzene(e)	Gas, Oil	Turbine	1-6	18,000	Ormat Turbines	Generator	2000	18(10 ⁶) hrs.	200	Some decomposition, slight corrosion if exposed to atmospheric humidity	Worldwide	75
Trichloroethylene (f)	Geothermal	Turbine	50	6,600	Instituto di Macchine	Generator	1	Initial operation in Nov. 79	70	None	Milan, Italy	78
Tetrachloroethylene (g)	Solar, Geothermal	1-Stage reaction turbine	4	1,200	Instituto di Macchine	Water pump Generator	1 4	1000 hrs. for all	80 80	Maintenance of condenser vacuum	Genoa, Italy Milan, Italy Ottana, Sardinia	46,78
Tetrachloroethylene (g)	Exhaust gases	Turbine	40	6,700	Instituto di Macchine	Generator	1	300 hrs.	115	Precautions to limit fluid temperature	Milan, Italy	78
FLUTEK PP3	Solar	Turbine	45	6,700	Instituto di Macchine	Generator	1	Initial operation in late 79	280	High cost of fluid	Milan, Italy	78
Alcohol/water	Gasoline	Multivane positive displacement	7.5	1,800	General Electric	Generator, dynamometer	None at present	16,000 hrs.	340	Some fluid decomposition and corrosion	Valley Forge, PA	71

(a) Experimental unit - simulated solar input.

(b) Laboratory testing.

(c) R-114 has been found to be the most compatible fluid for use as a coolant in the uranium enrichment gaseous diffusion process.

(d) Value estimated by author. Actual value is classified.

(e) C₆H₃Cl₃.(f) C₂HCl₃ (R1120).(g) C₂Cl₄.

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