SCREENING ANALYSIS FOR CHLORINE-FREE ALTERNATIVE REFRIGERANTS TO REPLACE R-22 IN AIR-CONDITIONING APPLICATIONS

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ABSTRACT

The potential health and environmental effects of the depletion of stratospheric ozone from refrigerants containing chlorine have resulted in international treaties, laws, and nonbinding agreements to phase out and eliminate many common refrigerants. R-22 is one of these compounds. A study was conducted to evaluate the potential of 22 chlorine-free compounds in refrigerant mixtures of up to three components as substitutes for R-22. The selection or screening of blends was based on vapor compression cycle COP at the 95°F (35°C) cooling condition, the volumetric cooling capacity of the blend, evaporator and condenser temperature glides, and the "estimated" flammability of the blend. Promising results were obtained for nine ternary blends containing E-125 and eleven ternary blends that exclude E-125. Recommendations are made to obtain further experimental data on E-125 since the mixtures with the best performance contain that compound. Results from this study will be used in an in-depth follow-on analysis.

INTRODUCTION

The role of chlorine in the destruction of stratospheric ozone and the resulting health and environmental risks have led to the Montreal Protocol to Protect the Stratospheric Ozone Layer, the London Amendments to the Protocol, and the U.S. Clean Air Act and the eventual total phaseout of chlorofluorocarbon (CFC) and hydrochlorofluorocarbon (HCFC) refrigerants. Although a great deal of work has been done to identify and develop replacements for R-11 and R-12 in centrifugal chillers, household refrigerators, and automotive air conditioners, relatively little effort to date has been directed toward finding substitutes for R-22 in heat pumps and residential air conditioners (Radermacher and Jung 1993). This paper reports on a project to identify blends of refrigerants that meet or exceed the performance of R-22 based on cycle efficiency, volumetric capacity, operating pressures, low ($\leq$10 F° [5.6 C°]) temperature glide, and flammability from a very large group of potential refrigerant mixtures.

REFRIGERANTS, BLENDS, AND COMPOSITION

Twenty-two different refrigerants (Table 1) were selected for analysis as possible components in binary and ternary blends to replace R-22. These refrigerants were chosen based primarily on their normal boiling points, although the selection criteria also included whether data are available for their critical temperatures and pressures. Some, like R-134a, R-152a, and R-290, have been studied extensively and a large body of information is available for them. Others, like the fluorinated ethers (e.g., E-125, CE-225ca), are relatively new and untested materials. All of the compounds in Table 1 are chlorine and bromine free, so all of them have ozone-depleting potentials of zero. It is also important that replacement refrigerants have low global-warming potentials (GWP); the GWP of a compound depends on its infrared absorption spectrum and its atmospheric lifetime. Longer atmospheric lifetimes significantly increase the GWP and the environmental liability of a chemical species. Some measured and estimated atmospheric lifetimes for the fluids screened in this analysis are shown in Table 1.

Nomenclature—and the intended meanings of a few specific words and phrases—is a problem in discussing mixtures of refrigerants and particular aspects of the mixtures. Terminology can be confusing since there is no clear way of referring to mixtures containing two or three components at specific weight percents or compositions and mixtures of fluids without any particular regard to the amounts of each. This discussion uses the terms *blend* and *mixture* to refer to a combination of refrigerants without regard to the percentages of each in the total and the term *composition* to refer to a combination of refrigerants at fixed or specific weight percentages of each.

The compositions of ternary blends are frequently illustrated using equilateral triangles, as shown in Figure 1. Each vertex represents 100% of one component and 0% of the other two; each edge shows binary mixtures (0% of the component on the opposing vertex) with compositions inversely proportional to the distances from each vertex. The points along the right edge of the triangle in Figure 1 represent 100% of the first component (at the top vertex), a 90%/10% blend of the first and second components (0% of the third), 80%/20%, 70%/30%, etc., down to the lower right vertex, which is 100% of the second component. The only truly ternary blends, non-zero percentages of each component, are in the interior region. In this study, each of the ternary blends that can be formed from the 22 refrigerants listed in Table 1 was investigated and the weight percents of each component in each blend were varied in 10% increments as pictured in Figure 1.

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TABLE 1
Properties of Fluids Used in Screening Analysis

Fluid	Chemical Formula	Critical Temperature		Normal Boiling Point		Molecular Weight	Atmospheric Lifetime (yrs)
		(°F)	(°C)	(°F)	(°C)		
R-32	CF_2H_2	173.2	78.4	-61.0	-51.7	52.02	6.1
R-125	CHF_2CF_3	151.3	66.3	-55.4	-48.6	120.03	28.1
R-143a	CF_3CH_3	163.6	73.1	-53.2	-47.3	84.04	41.0
R-290	C_3H_8	206.0	96.7	-43.7	-42.1	44.09	?
E-125	$CF_3\text{–O–}CF_2H$	177.3	80.7	-43.5	-41.9	136.02	21
R-218	$CF_3CF_2CF_3$	161.5	71.9	-38.2	-39.0	188.03	> 1000
CE-216	$\text{–}CF_2CF_2\text{–O–}CF_2\text{–}$	191.1	88.4	-19.1	-28.4	166.02	—
R-134a	CH_2FCF_3	214.0	101.1	-15.1	-26.2	102.03	15.5
R-152a	CHF_2CH_3	236.4	113.6	-12.4	-24.7	66.10	1.7
E-143a	$CF_2H\text{–O–}CFH_2$	220.8	104.9	-10.8	-23.8	100.04	3.4
R-134	HCF_2CF_2H	246.1	118.9	-3.7	-19.8	102.03	15.5
R-245cb	$CF_3CF_2CH_3$	227.3	108.5	-0.9	-18.3	134.04	1.8
R-227ca	$CF_3CF_2CF_2H$	223.3	106.3	2.7	-16.3	170.00	15
R-227ea	CF_3CFHCF_3	218.3	103.5	4.6	-15.2	170.03	30
E-227ca	$CF_3\text{–O–}CF_2CHF_2$	238.4	114.7	26.4	-3.1	186.03	—
R-236cb	$CF_3CF_2CFH_2$	266.2	130.1	29.4	-1.4	152.04	3.2
R-236fa	$CF_3CH_2CF_3$	267.1	130.6	30.0	-1.1	152.04	6.4
R-254cb	$CF_2HCF_2CH_3$	295.0	146.1	30.6	-0.8	116.06	1.6
CE-225ea	$\text{–}CF_2CHFCF_2\text{–O–}$	277.2	136.2	38.2	3.4	148.03	—
R-236ca	$HCF_2CF_2CF_2H$	282.1	138.9	41.1	5.1	152.04	—
E-134	$HCF_2\text{–O–}CF_2H$	296.8	147.1	43.1	6.2	118.03	2.8
R-236ea	CF_3CFHCF_2H	267.1	130.6	43.7	6.5	152.04	1.2

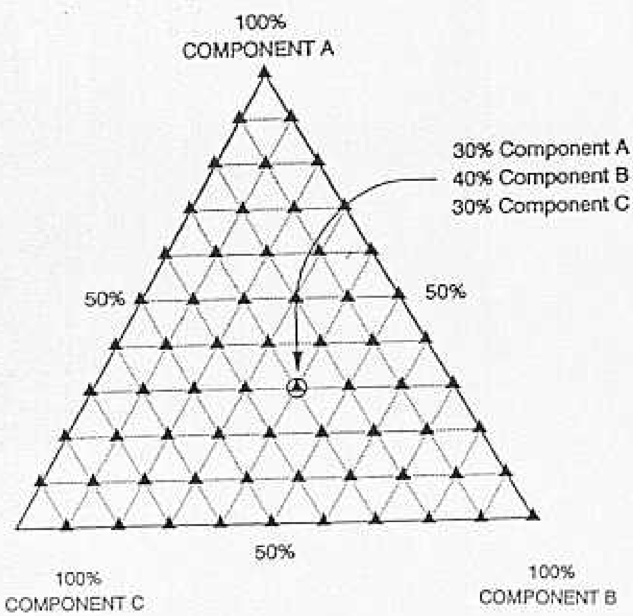

Figure 1 *Compositions of three components.*

This exhaustive approach to the calculations resulted in a very large number of compositions. The number of possible blends can be computed using Equation 1 where N= the number of pure fluids considered as components for a mixture and k= the number of components in each blend (k=3 for ternary mixtures, k=2 for binary mixtures):

$$\text{number of blends with } k \text{ components} = \frac{N!}{(N-k)! \times k!} \qquad (1)$$

Consequently, there are 1,540 unique ternary blends that can be formed using the 22 refrigerants listed in Table 1, and for each of these blends, calculations were performed for the 36 unique compositions in the interior of Figure 1. There are also 231 unique binary blends (0% of the third component) with 9 different compositions of each and, of course, 22 pure refrigerants to be considered. All told, then, 57,541 different compositions were evaluated in this study (1540×36 + 231×9 + 22). The large volume of information coming out of these calculations was stored in a data base for screening and evaluation.

LEE-KESLER-PLÖCKER EQUATION OF STATE

Computer calculations were used in this study to reduce the huge number of possible blends and compositions to a manageable number for more detailed study and experimental evaluation. The Lee-Kesler-Plöcker (LKP) equation of state was chosen for calculating refrigerant thermodynamic properties primarily because it is a corresponding states method (Plöcker 1977; Plöcker et al. 1978; Reid et al. 1986). The importance of this is that very little experimental data are required for each pure fluid in order to make reasonably accurate calculations of saturation and superheated vapor properties. In this case, the only information required for each of the 22 refrigerants listed in Table 1 are

- the critical temperature and pressure,
- the molecular weight,
- an acentric factor that is a measure of the acentricity or nonsphericity of the refrigerant molecules,
- correlations for the saturation temperature as a function of the reduced pressure (pressure divided by the critical pressure) and the saturation pressure as a function of the reduced temperature (absolute temperature divided by the critical temperature).

In fact, the correlation for saturation pressure is only used to find a starting point for an iteration, and an adequate correlation can be derived for the saturation pressure if one is known for the saturation temperature. Extensive effort has been made during the course of other projects to validate the results of calculations using the LKP equation of state. These are documented in Sand et al. (1991), Fischer and Sand (1990), and Fischer (1992).

There were two other principal reasons for the selection of the LKP equation of state for this study. First, computer subroutines had been obtained from Plöcker (1977) and Kruse and Kauffeld (1989) that were readily adaptable for use in this project and, second, a correlation was built into the subroutines for computing an interaction coefficient for binary pairs of pure components (Fischer and Sand 1990). This latter feature was virtually essential to the calculations in this study because of the extremely small amount of information available on some of the 22 refrigerants. Wherever possible, however, this correlation was supplemented with interaction coefficients determined by experimental data.

CYCLE CALCULATIONS

An important simplification was made in the refrigeration cycle analysis in order to reduce the magnitude of the computations performed. McLinden and Radermacher (1987) showed that the results of ideal cycle calculations for mixtures are highly dependent on the assumptions for how the refrigerant-side conditions are determined. The preferred calculation is one where heat transfer fluid temperature differences and total heat exchanger loading are specified instead of refrigerant-side temperatures. This approach, however, requires iterations that increase the computational overhead to a point that was deemed unacceptable for the 57,000 blends considered in this screening study. Even an evaluation using specified mid-point temperatures in the heat exchangers requires an iteration that was considered excessive in this analysis. Consequently, this paper presents intermediate results from a simplified calculation; as such, it serves a primary purpose of reducing the number of blends to consider for further analysis. Refrigerant temperatures were selected that are appropriate for a high-efficiency air conditioner at a 95°F (35°C) ambient air temperature. These are

- 120°F (48.9°C) dew-point temperature entering the condenser,
- 50°F (10.0°C) dew-point temperature leaving the evaporator,
- 60°F (15.6°C) return gas temperature (10 F° superheat), and
- 105°F (40.6°C) liquid line (15 F° subcooling for R-22 and less subcooling for gliding mixtures).

An approach such as this that relies on dew-point temperatures will give slightly higher values for calculated COPs and lower values for capacity than calculations based on mean heat exchanger temperatures (McLinden and Radermacher 1987). The low-glide mixtures identified by this screening as attractive alternatives to R-22 need to be evaluated at both the heating and cooling rating conditions using a rigorous cycle model that can do a more accurate simulation of changes in heat exchanger and compressor performance resulting from gliding refrigerant temperatures during evaporation and condensation (Domanski and McLinden 1990; Rice and Sand 1990; Jung and Radermacher 1991a, 1991b; Radermacher and Jung 1993).

The simulated performance of R-22 at these conditions is 122 Btu/ft^3 (4550 J/L) volumetric cooling capacity with a COP of 6.30 (100% isentropic compression). Quantities resulting from the calculations are

- the COP and volumetric cooling capacity at 95°F (35°C),
- the high- and low-side refrigerant pressures,
- the liquid-line and return gas specific volumes, and
- the flammability index[1] ($FI<0$ for nonflammable blends, $FI>1$ for flammable blends, and $0 \leq FI \leq 1$) for indeterminant flammability.

The calculated values for each mixture are stored in a large (five-megabyte) data base for subsequent screening and

[1] "Flammability index" is an undocumented parameter derived from confidential business information provided as a courtesy by a refrigerant manufacturer.

evaluation. Data are filed for *all* blends and composition without any pre-screening to reduce the magnitude of information. The flammability index is the "softest" quantitative property calculated for each mixture, and any conclusions based on it must be supported with laboratory tests on the limits of flammability.

RESULTS

The calculated performance information in the computer data base was screened to identify nonflammable compositions that have predicted COPs and capacities close to or greater than that of R-22 with temperature glides less than 10 F° (5.6 C°). Many of these could be grouped as different compositions of mixtures of the same three components; these data were plotted on triangular diagrams as shown in Figures 2 and 3 for graphical evaluation. Figure 2 indicates the regions of flammable (heavily shaded) and nonflammable (lightly shaded) blends of R-32, R-134a, and R-227ea (a region of indeterminant flammability is also indicated). This example also shows contours of constant COP at 95°F (35°C) ranging from 98% to more than 132% of the COP of R-22. There is also a single bold contour where the cooling capacity is the same as that for R-22. Figure 2 shows that there are combinations of R-227ea and R-32 where the capacity is nearly the same as that of R-22 with COPs as much as 32% higher (along the left edge). What it does not show is that there is a large evaporator glide for these compositions and that the compositions with evaporator glides of less than 10 F° (5.6 C°) are all either in a narrow band along the bottom of the triangle in Figure 2 or up in the flammable region. Figure 3 displays results for mixtures of R-32, R-125, and R-134a and is similar to Figure 2 except that it also includes diagrams showing contours for cooling capacity, high-side pressure, and evaporator temperature glide. It also identifies five different mixtures that are being considered by other investigators as R-22 replacements. Graphs of this type were used to select the "best" blends for future in-depth evaluation.

This preliminary screening identified 13 blends with temperature glides of less than 10°F at some composition (i.e., specific weight percents of each component) that are attractive with regard to efficiency, capacity, operating pressure, and flammability.

- R-143a / E-125 / R-227ea
- R-32 / CE-216 / E-143a
- R-32 / E-125 / R-218
- R-32 / E-125 / R-227ca and R-32 / R-125 / R-227ca
- R-32 / E-125 / R-227ea and R-32 / R-125 / R-227ea
- R-32 / E-125 / R-245cb
- R-32 / E-125
- R-32 / R-125 / R-134a
- R-32 / E-125 / R-134a and R-32 / E-125 / R-134
- R-32 / E-125 / CE-216

In some cases both COP and capacity are predicted to be higher than those for R-22.

The frequency of the R-32/E-125 pair in these promising blends emphasized the need for resolving any uncertainties about the toxicity of E-125 (Simons et al. 1977) and for additional physical property measurements on this potentially useful refrigerant (Wang et al. 1991). Laboratory data,

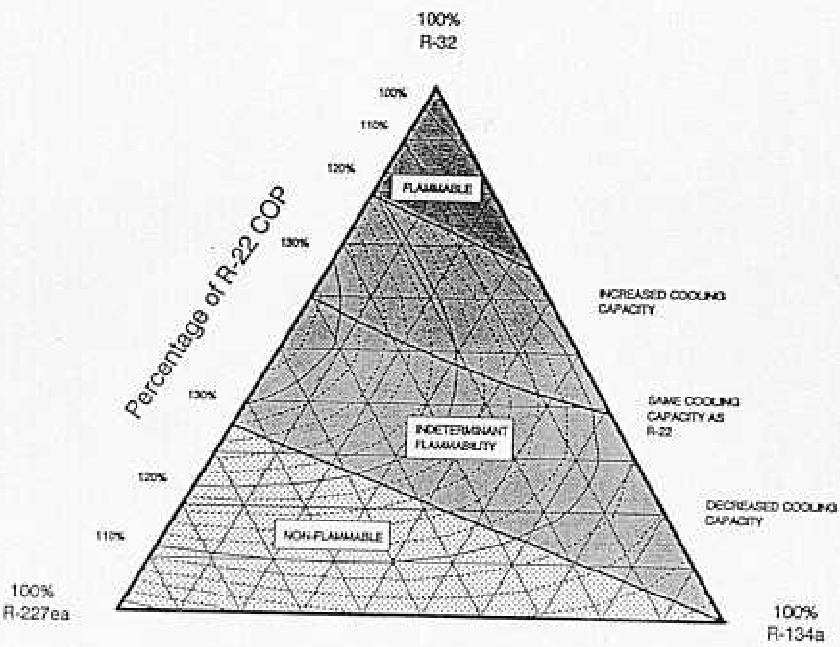

Figure 2 *Lines of constant COP relative to R-22 for mixtures of R-32, R-134a, and R-227ea with flammable and nonflammable regions of increased and decreased capacity.*

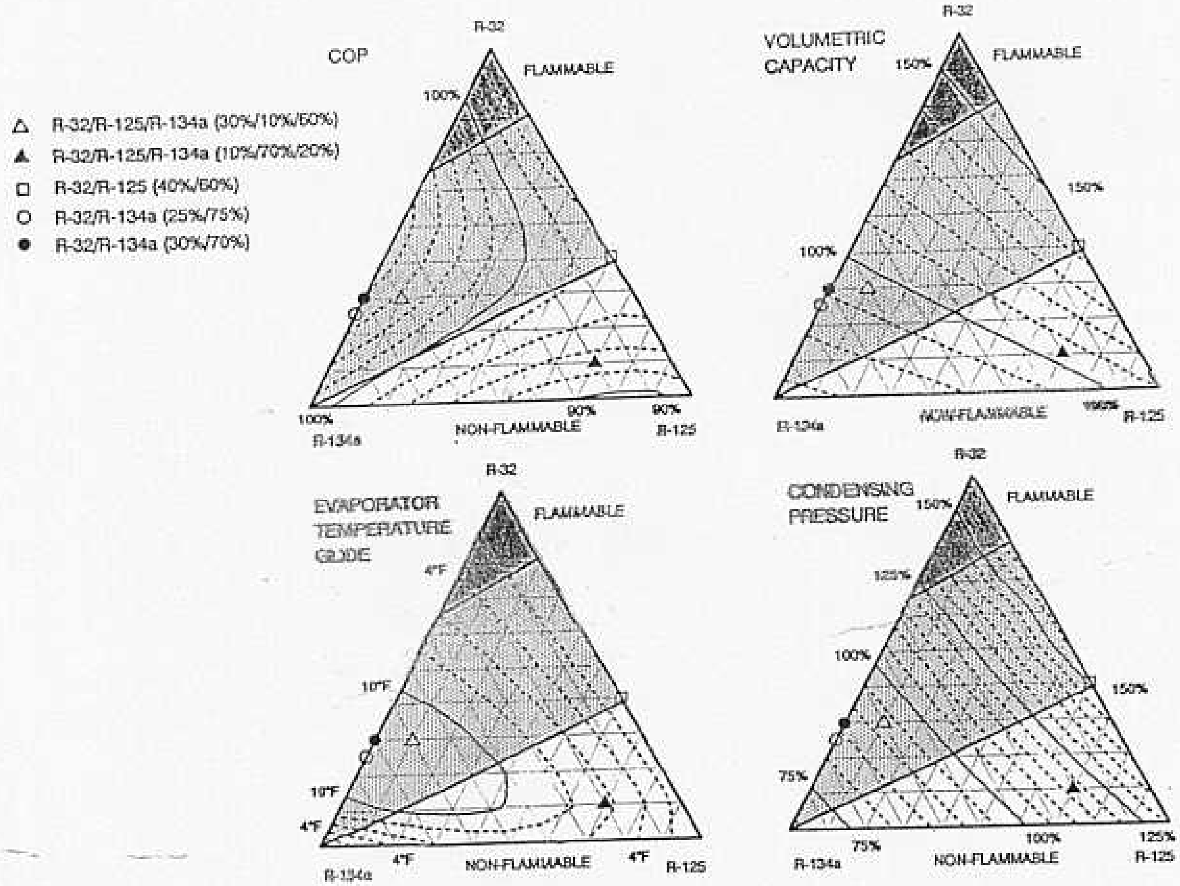

Figure 3 *COP, cooling capacity, evaporator temperature glide, and condensing pressure for mixtures of R-32, R-125, and R-134a.*

particularly measurements that can be used to calculate interaction coefficients for E-125 with other refrigerants, would be extremely valuable in refining these predictions.

Other attractive blends excluding E-125 from the composition include:

- R-143a / R-218 / CE-216
- R-143a / R-218 / R-134a
- R-32 / CE-216 / R-134
- R-32 / R-125 / E-143a
- R-32 / R-125 / R-245cb
- R-32 / R-134a / R-134
- R-125 / R-143a / R-218

In general, these seven blends have lower efficiencies, higher pressures, or are more likely to be flammable than those listed in the first set of blends. Table 2 lists some general observations for "feasible compositions" (i.e., less than 10 F° [5.6 C°] glide, nonflammable) of 18 of the 20 blends listed above (the R-32/E-125 binary is excluded because it is covered by the ternaries including R-32 and E-125; the one blend containing R-218 is excluded because of computational problems like those described in the next section).

COMPUTATIONAL PROBLEMS

The brute force approach toward the screening analysis, where tens of thousands of calculations are performed exhaustively without a great deal of forethought about the practicality of evaluating some blends, will almost inevitably run into some computational problems. In this particular analysis, "unresolved errors" occurred in almost 1,500 of the more than 57,000 compositions evaluated. These problems were experienced very early in the project, and a conscious decision was made to keep track of which compositions were not evaluated successfully but not to put forth any effort at this time to correct the problems.

A cursory examination of a few blends with computational problems revealed two sources of numerical difficulties, both of which are illustrated in Figure 4. This figure shows the cycle diagram for a high-efficiency air conditioner superimposed on the p-h diagram for R-218. In this case, the vapor dome is tilted sufficiently that the specified operating conditions result in two-phase compression, an obvious problem. Although it would have been possible to adapt the cycle calculations to accommodate or avoid this kind of situation, these modifications were not made. Changing the computer programs to handle two-phase

TABLE 2

Selected Refrigerant Blends That Have Potential Applications as R-22 Substitutes Due to Low Temperature Glides, Comparable or Better Cooling COP and Capacity, and Predicted Nonflammable Compositions

First Component	Second Component	Third Component	Observations
1. R-32	R-125	R-134a	up to 4% increase in COP with up to 20% increase in capacity; broad region of feasible compositions
2. R-32	E-125	R-134a	6 to 8% increase in COP with up to 30% increase in capacity; broad region of feasible compositions with lower glides than R-32/R-125/R-134a
3. R-32	E-125	R-134	7 to 10% increase in COP with 10 to 30% increase in capacity; higher glides than R-32/E-125/R-134a
4. R-32	CE-216	R-134	very few non-flammable compositions with low glides; extremely small non-flammable region; -10 to +6% change in COP with 20 to 30% decrease in capacity
5. R-32	R-125	E-143a	2 to 4% decrease in COP with 10 to 50% increase in capacity
6. R-32	R-125	R-245cb	very few feasible compositions; generally high temperature glides; -4 to +2% change in COP with 10 to 50% higher capacity
7. R-32	R-134a	R-134	generally high temperature glides; low concentration of R-32 essential for non-flammability; high loss in capacity with loss in COP
8. R-32	E-125	CE-216	6 to 20% increase in COP with -10 to +30% change in capacity; almost all compositions have less than 10°F glides; broad region of feasible compositions
9. R-32	E-125	R-245cb	6 to 12% increase in COP with 10 to 30% increase in capacity
10. R-32	E-125	R-227ea	up to 20% increase in COP with up to 30% increase in capacity
11. R-32	E-125	R-227ca	5 to 20% increase in COP with up to 30% increase in capacity
12. R-32	R-125	R-227ca	up to 10% increase in COP with 10 to 20% increase in capacity
13. R-32	CE-216	E-143a	no clearly non-flammable ternaries; up to 10% increase in COP with at least a 40% loss in capacity
14. R-143a	E-125	R-227ea	±4% change in COP with up to 15% loss in capacity; COP increase corresponds to capacity decrease
15. R-143a	R-218	CE-216	losses in COP and capacity relative to R-22; up to 30% loss in capacity and 4% loss in COP
16. R-143a	R-218	R-134a	2 to 4% loss is COP with 5 to 25% loss in capacity; broad range of feasible compositions; all compositions have glides less than 10°F
17. R-32	R-134a	R-227ea	2 to 6% increase in COP with large decrease in capacity
18. R-32	R-125	R-227ea	-2 decrease to 8% increase in COP with 20 to 50% increase in capacity

compression would have increased execution time for all 57,000 compositions evaluated in order to avoid problems with a small percentage of cases. The problem blends were noted so that they can be handled individually if they seem to merit further attention.

The other difficulty illustrated in Figure 4 appears where the top of the vapor dome has been lopped off. The computer subroutines obtained from Kruse and Kauffeld (1989) imposed limits so that saturation properties could not be calculated for conditions "close to" the critical temperature, "close" being within 90% or 95% on an absolute scale. This limit was imposed because of the poor performance of the LKP equation of state close to the critical temperature, and the judgment of the original authors of those subroutines was accepted and used in this analysis.

Figure 5 summarizes information on the blends of refrigerants that experienced computational difficulties. There is a triangle plotted for each of the 22 refrigerants considered in the study. The horizontal axis is the critical temperature for each refrigerant, and the vertical axis is the number of compositions containing that refrigerant that had computational errors. Several of the points are also labeled with the corresponding refrigerant numbers. There is an almost obvious connection between "low" critical temperature and computational difficulty indicated by the sharp break between the points in the upper left of the graph and those in the lower right. Although this figure seems to explain some of the problems, it also poses some unanswered questions: why do CE-216 and R-134a have less than half the number of problems than compositions

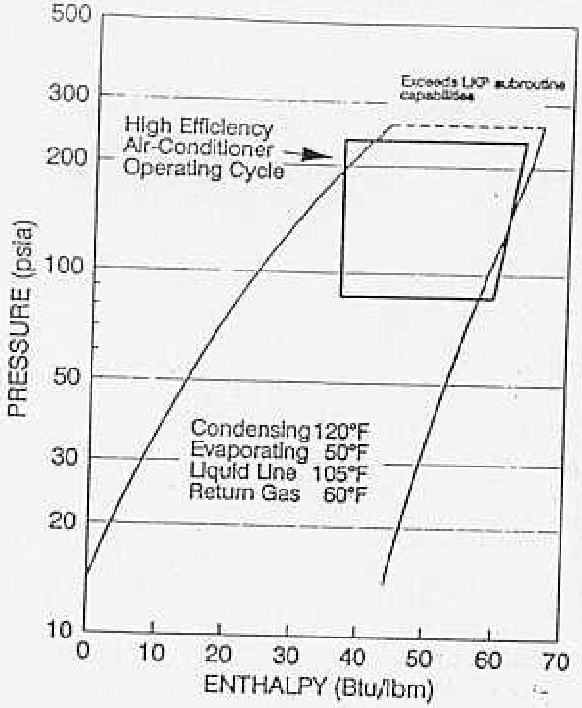

Figure 4 *Pressure-enthalpy diagram for R-218 with superimposed air-conditioning cycle.*

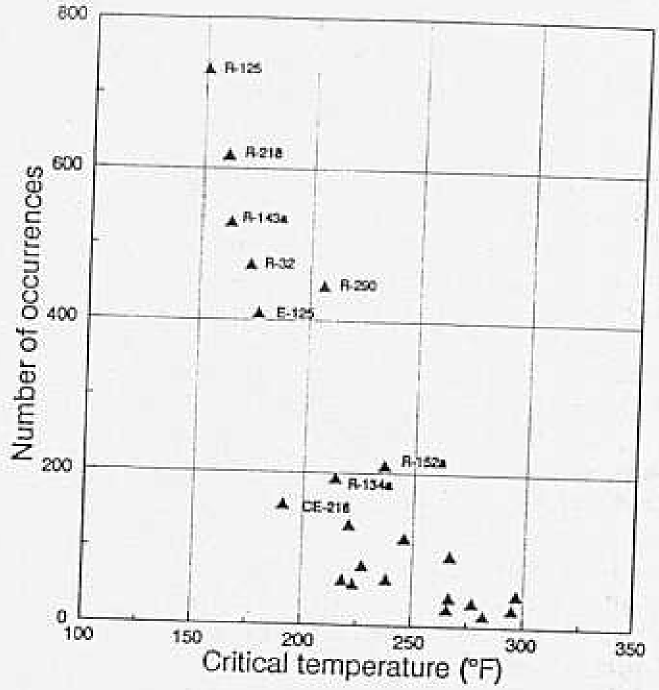

Figure 5 *Occurrences of computational problems in the R-22 screening analysis and refrigerant critical temperatures.*

containing R-290, which has a higher critical temperature? Interestingly, only one of the nearly 1,500 compositions with computational errors does not contain any of the six refrigerants in the upper left of Figure 5 (i.e., R-125, R-218, R-143a, R-32, R-290, and E-125) and only 241 compositions contain just one of those six refrigerants. The other compositions contain two or more of the six refrigerants with "low" critical temperatures.

As mentioned earlier, it is possible to put further effort into examining and correcting the computational problems with some or all of these blends, although it would be prudent to focus attention on a select few. Of particular interest would be compositions containing R-32 and R-218 since one fluorocarbon manufacturer is interested in an azeotrope of these refrigerants. Effort toward understanding the problems with R-290 would also be worthwhile since this refrigerant is mentioned by another manufacturer as a component of quaternary blends it is studying. None of the other "problem" blends appear interesting at this time.

CONCLUSIONS AND RECOMMENDATIONS

Several promising blends of chlorine-free refrigerants with low dew-point to bubble-point glides have been identified as potential alternatives for R-22. Several of these refrigerant mixtures contain E-125, one of the components that highlight the necessity of additional physical property and toxicity measurements for this refrigerant. Many of the potential alternatives contain hydrofluorocarbon (HFC) components that are scheduled for commercial production.

Further analytical studies are planned for interesting blends arising from this broad-based screening. Initially, more elaborate computer models will be used that are better for simulating Rankine cycle performance at heating and cooling conditions other than 95°F (35°C) cooling. Additionally, these models can also emulate heat exchanger performance with pure and zeotropic refrigerants more accurately.

System performance tests, where these blends are used as "drop-in" replacements for R-22 in existing equipment or as R-22 alternatives in breadboard loops where operating conditions and circuit configurations can be more easily modified to suit individual refrigerant characteristics, are required to support these analytical results. As with R-134a, transport properties could have an important influence on relative refrigerant performance. Laboratory testing in a breadboard test loop is planned for several of the more efficient and accessible blends.

Additional analytical screening is planned for refrigerant blends with larger refrigerant glides in the heat exchangers (> 10°F; 5.6°C). Zeotropic refrigerants with larger gliding temperatures can be used to improve cycle efficiencies compared to pure refrigerants if appropriate hardware modifications are incorporated into the vapor compression system (Kauffeld et al. 1990).

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