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COST REDUCTIONS IN ABSORPTION CHILLERS

PHASE II

R.W. Leigh, Principal Investigator
Contributors: H.S. Isaacs, J. Kirley,
J.G. Murray, A. Ravve

February 1989

Prepared for the
GAS RESEARCH INSTITUTE
8600 West Bryn Mawr Avenue
Chicago, IL 60631

DEPARTMENT OF APPLIED SCIENCE

BROOKHAVEN NATIONAL LABORATORY
UPTON, LONG ISLAND, NEW YORK 11973

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0. INTRODUCTION AND SUMMARY

0.1 Introduction

The absorption chiller market in the United States is in deep trouble, with sales of less than 100 domestic and 100 Japanese units in 1984 [0-1]. The problem is essentially that the compression alternative is inexpensive, reliable and efficient enough to offer acceptable operating costs. It appears that absorption chillers, and the gas markets that they will create, can only be resuscitated by substantial improvements in their salient characteristics, whether these be lowered first costs, or improvements in efficiency.

A research program at Brookhaven National Laboratory (BNL) [0-2] has addressed the possibility of dramatically lowering the first costs of absorption chillers through lowered material intensity and the use of lower cost materials, primarily in the heat exchangers which make up the bulk of the operating components of these systems. This must be done while retaining the best performance characteristics available today, a gross design point coefficient of performance (COP) of 1.3 and a net design (seasonal) average COP of 1.0 (0.90) in a directly fired, double effect unit.

We have investigated several possible routes to these goals, and here report on these findings, focusing on the areas that appear most promising. The candidate technologies include the use of polymer film heat exchangers in several applications, the use of thin strips of new, corrosion resistant alloys to replace thicker, less impervious metals in applications exposed to gas flames, and copper or cupro-nickel foils in contact with system water. The use of such materials is only possible in the context of new heat exchanger and system designs, which are also discussed.

To lend focus, we have concentrated on a directly fired double effect system providing capacity only. If successful, these techniques will also find wide applicability in heat pumps, cogeneration systems, solar cooling, heat recovery and chemical process heat transfer.

0.2 Background

Our cost goal approach originated in work done for the U.S. Department of Energy in pursuit of solar energy systems which could compete economically with conventional energy sources [0-3]. An assessment demonstrated that appropriate retail costs were far below those then currently available, forcing a significant re-design of solar collectors. The key to the new designs was lowered material intensity, since the cost goals dictated that only very small amounts of anything that was not free (air, for example) could be included in the collector. Thin films of material, both polymers and metal foils, were identified as likely candidates, and the problem at that point was reduced to the engineering problem of finding materials which could perform adequately while withstanding the rigors of the outdoor environment.

Absorption chillers are ripe for a similar analysis and development. While there are many advantages to the underlying design, including quiet, reliable operation, reasonable resource use efficiency and the possibility of cooling through the recovery of cogenerated heat, the systems are currently too expensive to compete with the electric alternative.

In Phase I of this project we therefore analyzed the economic circumstances of absorption chillers to determine cost goals both for complete systems and on a component by component basis. We determined, for example, that a 25 ton machine should compete well with vapor compression devices in areas of high electricity prices if it could be sold for \$9000. We then examined available information on possible thin materials to select an initial set of candidates which would meet both technical and cost requirements. Information on the interactions of many materials with the lithium bromide brine used in the chillers was sparse or non-existent, so we report on a set of experiments which allowed us to begin the process of winnowing out the candidates least able to tolerate the environment to which they would be exposed.

We also established baseline designs for assessment purposes. These are systems which could be built with the best of current industry practice and are referred to in this Phase II report as "BCP" chillers. In these baseline designs for systems of 3, 25, 100 and 400 tons we developed tables delineating both the operating conditions which are faced by heat exchangers in these machines and the current practices used in constructing them, including materials choices, flow rates, temperatures, pressure differentials and other information relevant to the assessment of heat exchanger materials; the tables for the 25 ton machine are reproduced here as Appendix 2. Finally, to demonstrate that chillers could be constructed using these materials, we developed conceptual designs. The results of this work are reported in Ref. 0-1, and serve as the basis of the second phase of this effort, which is reported here.

0.3 Summary of Results

Polymer films were a prime candidate because they are impervious to corrosion. We screened a large number of materials, especially for thermal conductivity and tensile strength. We tested over two dozen of the most promising candidates for short term tensile strength at room temperature and at 93°C (200 °F). We then measured the creep under stress and the durability of about six of the most promising of these for one year in 105°C (220 °F) lithium bromide brines; several candidates performed satisfactorily, the most outstanding being polyether-ether-ketone (PEEK), which was also tested at 150 °C (300°F) and may also be a viable candidate even at that temperature. We then analyzed the various heat exchangers in our BCP chillers and determined that the second effect generator and one or both recuperators were viable candidates to be manufactured from PEEK (or possibly PFA Teflon), but that the permeability of all polymers rendered them unsuitable for the evaporator, absorber or condenser, since they would allow oxygen into the chiller from the circulating water.

We also constructed a small shell and tube recuperator from PEEK and tested its performance in (approximately) counterflow heat exchange. Performance, displayed below on a Wilson plot, was poor due to the small size of the device but sufficient to indicate that polymer recuperators constructed using design criteria developed for comparable metallic heat exchangers will perform in accordance with standard engineering expectations. The experience of construction led us to the conclusion that the best approach to fabrication, especially manifolding, would be blow molding from a pre-welded

flat intermediate structure. The welding of the detailed structure is especially adaptable to automation.

We also developed methods for coating thin foils of various metals with an electroplated epoxy plating and tested three different coatings for integrity, tenacity and longevity in brines. We found it possible to lay down fault-free, uniform coatings on bare and tinned steel, copper and aluminum which would in the short term prevent any contact between fluids and the metal. We were able to select one coating as best on the basis of flexibility, a key item on these thin foils. The coating were some 0.7 mils thick, and so might have been acceptable with respect to heat transfer, had we progressed to heat exchanger construction. However, several problems were uncovered. One negative finding that emerged was our inability to coat the interior of any complex object, such as the inside surface of a pipe or heat exchanger; this appears to be an intrinsic aspect of electro-deposition which must simply be designed around. More devastating were the results of long term immersion tests in open (oxygenated) lithium bromide brines at 220°F: although many samples looked quite good after about two months of soaking, at the end of six months the coatings had become brittle and usually separated from the substrates. It appears that this widely used epoxy is incompatible with the brine at these temperatures. An attempt to identify other (non-epoxy) coatings should be a part of future work. Since it was clear that these coatings were unsuitable, we made no attempt to measure their heat exchange properties.

We constructed an instrumented counterflow heat exchanger test rig, which was used here to test the PEEK recuperator. The system remains available for future use.

Thin metal foils can also be used to lower material intensity, but the lack of adequate fabrication techniques impedes implementation since welding techniques are critical to making heat exchangers. We had previously developed an ultrasonic welding facility which has been used very successfully on coppers in other applications. We can also weld aluminum and nickel alloys easily and reliably. Steels have historically been a problem, since the weld head is steel and tends to stick to the material. (The other options for thin materials, laser welding and electron beam welding, are undesirable because they are quite expensive, and electron beam welding is not reliable for our very thin foils.) After considerable experimentation and analysis, we have demonstrated the consistent and reliable ultrasonic seam welding of stainless steel foils using a proprietary aluminum oxide coating on the steel weld head. This development has the potential for many applications in areas other than absorption chillers or even heat exchangers. We were unable to extend the technique to bare or tinned steel foils, which seem to harden to the point of extreme brittleness under the action of the weld head.

The use of thin metal foils is also constrained by the dangers of corrosion, made more severe by the imminent danger of penetration of these thin materials. We therefore continued our work from the first phase to determine the corrosion mechanisms most likely to occur in alloys used in a thin materials chiller and to identify materials which would make possible the construction of such a chiller. In view of the limited time available for this study, rapid testing was carried out where virtually no information was available from the literature.

Copper and steel have been used successfully in chillers and data on the behavior of carbon steel, types 304 and 316 stainless steels, nickel, cupronickels and Monel is available. In general, most metals, including copper, corrode uniformly in LiBr. The use of superalloys should reduce the corrosion rates below that of stainless steel if they are not susceptible to various forms of localized corrosion, since stress corrosion cracking can be prevented by increasing the pH of the solutions. Measurements were therefore carried out to determine the susceptibility of a series of superalloys to localized corrosion. As a result of these tests we have established that several superalloys are suitable for use in the first effect generator with respect to localized corrosion, especially AL6-X when cost is taken into account.

Crevice corrosion was also considered a possible mode of corrosion of ultrasonically welded copper in the coolant waters. The narrow gaps present along the welds form tight crevices and if crevice corrosion could take place this would limit the life of the copper heat exchangers. There was no crevice corrosion information available for copper, and as the metal is susceptible to pitting in certain environments accelerated measurements were carried out on copper crevices to determine their corrosion characteristics. From these measurements we believe that crevice corrosion will not be a problem for copper foils, although there is still a possibility of pitting corrosion in these components which must be examined at a later date.

Our proposal to make the second effect generator from PEEK raised the question of how well a polymer heat exchanger would perform in boiling heat transfer, since its smooth, low surface energy interface might inhibit nucleation. We therefore constructed a simple instrumented boiler from a bag made of PEEK and filled with heat transfer oil and a resistance heater, with a ladder of thermocouples inside and outside the bag. The bag was immersed in water and heated. The results, described in detail in Chapter 9, are encouraging. Many nucleation sites were involved in boiling, with their number increasing with heat transfer rate, and the observed temperature differences across the interface were consistent with efficient generator operation.

The broader question of the enhancement of heat transfer during phase change processes is addressed in Chapter 10. By reviewing the literature on process experiments and equipment demonstrations, we can conclude that we can expect substantial benefits from the use of PEEK or other polymers wherever condensation occurs, since that condensation will be drop-like under a wider range of conditions than for metals; this will result in specific enhancement of the performance of a second effect generator over the performance discussed in the previous paragraph.

We also prepared a computer simulation model with which we assessed the overall feasibility of our baseline design and determined the performance impact of design choices. We obtained a copy of recently completed "ABS" model developed by Dr. G. Grossman and calibrated it by successfully representing the "best current practice" design for a 25 ton chiller developed during Phase 1 of this effort. We then adjusted the parameters representing the sizes and connections between the various heat exchangers to match the requirements of our conceptual design, based on concentric cylinders. Under a variety of qualifying assumptions, we are able to predict from calculations using this model that this design would operate with a fuel-based COP of 0.99

(including stack losses) and a heat-based COP of 1.32. Chapter 11 also contains an illustrated description of our conceptual design, describing some of the functional relationships, assumptions and remaining undefined details.

Taking the conceptual design presented in Chapter 11 as a baseline, we then estimated the expected costs of producing a 25 ton chiller using thin materials technology. Much of the cost data is drawn from the final report on Phase I of this work [0-1], and, as there, we find a considerable amount of uncertainty inherent in the estimates. Nonetheless, a 25 ton thin materials chiller can probably be built for a total materials cost of \$3380, an estimated labor cost of \$3450, and sell, after markups, for a total equipment cost of about \$11,000, or \$440/ton. This is close to the breakeven cost goal established in our Phase I report of \$360/ton or a total of \$9000 for a 25 ton machine (12-3). It misses that goal because of costs not associated with heat exchangers, which may also be susceptible to cost reduction, but which were not specifically studied here. Thus we find that if cost reductions can also be achieved in these areas which have not as yet been investigated, there is a real chance of meeting this cost goal and establishing a marketable chiller.

1. POLYMER MATERIALS EVALUATION

1.1 Approach and Selection Criteria

This section presents our investigations to determine which thin polymer films could be used in each of the heat exchangers comprising an absorption chiller. Because a great deal of information on the properties of polymer films is tabulated and readily available, our first task was to collate existing data on polymer properties and build a data base which could be used for the initial selection process. This data base was then used to develop a broad set of candidates for initial testing. Initial testing measured the stress-strain characteristics of the candidates under relatively high stresses over a short period of time at 38 and 93 °C (100 and 200 °F). The few candidates scoring best in this test were then subjected to long term creep tests in Lithium Bromide brine at 100 °C (212 °F). The results of these tests were combined with information in the data base to pick out a narrower set of candidates (sometimes an "empty set") for each heat exchanger in the final selection process.

Limitations on available material strength data constrained the selection process. Unlike metals and other traditional materials, polymers are visco-elastic and have a complex response to steady imposed stress. In particular, the standard measures of yield strength (the lowest stress for which additional strain can occur with no increase in stress) and the ultimate tensile strength (the maximum stress observed on a stress-strain curve before the material begins to rupture or stretch uncontrollably) are only applicable over relatively short time periods. Polymers which are subjected to steady stress will also slowly stretch, or "creep", and this, rather than tensile strength, is the constraining design consideration. Data on long term creep is, however, much more expensive to obtain (due to the duration of the tests) and therefore available only for selected materials, and even then the data were for bulk materials rather than films. For the initial selection process then, we used tensile and yield strength data; only the tensile strength data were widely available for materials in film form. The creep tests carried out on the most promising materials and described in Section 1.5 were used as the basis of the final selection.

1.2 Polymer Film Data Base

This task consisted of the expansion of a data base developed under Phase I, drawing on the information gathered there, information available in more recent published reports (1-1,2,3,4), expert opinion from our consultant (Dr. A. Ravve) and the use of a commercial "on-line" computerized polymer data base ("Plaspec", ref. 1-5). An abbreviated version of the data base is presented in Table 1-1, and includes all polymers known to us to be available in film form (by which we mean a thickness of less than 10 mils). The core of the information is derived from the Modern Plastics Encyclopedia (1-1), augmented by manufacturer's data and expert opinions. The data are all based on standard tests prescribed by the American Society for Testing Materials. Blank spaces indicate that the information could not be found.

Table 1-1
Abbreviated Polymer Film Data Base

Brand Name	Acronym	Chemical Name	Tensile Strength psi	Elongation (%) Min Max	Water ¹ Abs 24hr %	-O ₂ ² Trans- mis- sion	Resis- tance Per- mea- bility	Thermal Cond. Btu-mil Hr-F-Ft ²	Melt Temp °F	Deflectn Temp(°F) psi	Resin Price cents/ ft ² -mil
Barex	ABS	Acetal Copolymer	8800	40 75	0.22		G G	1600	335	185 311	1.1
		Acetal Homopolymer	9500	13 75	0.4		G G	1600	350	255 328	1.1
		Acrylonitrile-butadiene-styrene	5000	10 50	1	70	FP G	1810	220	170 170	0.5
		Acrylonitrile methyl acrylate	9000	10 150	0.3	5 0.8	G G	870	400	165	0.7
		Cellulose Acetate	7000	15 55	8.5	40 150	P G	1740	445	111 120	0.9
		Cellulose Acetate Butyrate	5000	50 100	2	40 1000	P G	1740	284	113 130	0.8
		Cellulose Propionate	4000	60 80	2.5	890	P G	1740	374	111 147	0.8
		Cellulose Regenerated	7000	10 50	115	134 0.8		1740			
		Cellulose Triacetate	9000	10 50	4.5	40 150	FP G	1740			
		Ethyl Cellulose	8000	20 30	7.5	14.2 2000	P G	1570	275	115	
Halar	ECTFE	Ethylene Chlorotrifluoroethylene	8000	150 250	0.02	0.6 25	G G	1100	450	170 240	10.1
Tefzel	ETFE	Ethylene tetrafluoroethylene	7000	300 300	0.02	1.65 100	G G	1650	520	160 220	12.7
Teflon	FEP	Fluorinated Ethylene-Propylene	2500	300 300	0.01	0.4 750	G G	1750	530		158 11.0
Teflon	PFA	Perfluoroalkoxy	4000	200 600	0.03		G G	1750	590		166 21.2
Aclar	PCTFE	Polychloro-trifluoroethylene	5000	50 150	0 0.05	15		1450	430		258 34.3
Teflon...	PTFE	Polytetrafluoroethylene	1500	100 400	0		E E	1750	620		250 7.3
Tedlar	PVF	Polyvinyl Fluoride	7000	115 250	0.5	3.2 3	G G	1650			
Kynar	PVDF	Polyvinylidene Fluoride	5500	300 500	0.04	2.6 14	G G	800	350	188 280	6.0
		Ionomer	5000	250 450	0.4	2 400	G G	1780	191	93 113	0.6
Nylon 6		Polyamide - Biaxially Oriented	32600	100 120		11 1.4	G GP	1680	420	155 365	0.9
Nylon 6		Polyamide - Extruded	9000	250 550	9.5	22 2.6	G GP	1680	420	155 365	0.9
Nylon 6		Polyamide - Uniaxially Oriented	10000	60 450		15 2	G GP	1680	420	155 365	0.9
Nylon 11,12		Polyamide	7000	250 400	0.27	4 92	G G	2320	380	130 300	1.9
		Polybutylene	1700	300 380	0.01		G G	1510	130	215	0.4
Lexan...	PC	Polycarbonate	8400	40 105	0.35	11 300	GP G	1360	270	280	1.2
	PET	Polyethylene terephthalate	20000	60 165	0.8	1.3 6	G G	1000	490	70	1.2
	PEEK	Polyetheretherketone	13200	30 150	0.14			1420	633	320	16.8
Ultem	PEI	Polyetherimide	15200	10 60	0.25		G GE	460	421	383 398	2.8
Victrex	PES	Polyethersulphone	12200	6 80	1.7		GF GE	1100	437	394	3.0
	PE-LLD	Polyethylene (Linear Low Density)	1400	100 965				2320	253		0.2
	PE-LD	Polyethylene (Low Density)	1500	100 700	0.01	1.5 500	F G	2320	224	104	0.2
	PE-MD	Polyethylene (Medium Density)	2000	50 650	0.01	0.7 535	F G	2320	224	104	
	PE-HD	Polyethylene (High Density)	2400	10 650	0	0.3 185	G G	3340	272	175	0.2
	PE-HMW	Polyethylene (Hi Mol Wt)	2800	170 800				3340	266	154	0.2
	PE-UHMW	Polyethylene (Ultra Hi Mol Wt)	3000	300 300	0		G G	3340	266	110 155	0.5
	PE-COP	Polyethylene-vinyl acetate	1000	400 800	0.01	3 840	F G	1721	222		0.3
		Ethylene-methyl acrylate	1650	740	0		F	1885	181		
		Polyimide (Thermoplastic)	25000	70 70	2.9	5.4 25	G G	2460	640	530	
Kapton		Polyimide (Thermoset)	23000	20 50	4	5.4 25	G F	1250			
Acrylic	PMMA	Polymethyl methacrylate-stndrd	8200	4 12	0.4	1.3	G	1450	203	165 175	0.5
Korad Ac	PMMA	Polymethyl methacrylate-Type A,D	5100	75 75	1.6	8	PG G	1450			0.6
TPX		Polymethylpentene	2700	10 10	0.00		F E	1160	450	120 175	
	PP	Polypropylene - Extrusion	4500	550 999	0.00	0.7 240	G G	810	340	120 225	0.2
	PP	Polypropylene - Oriented	7500	35 475	0.00	0.4 160	G G	810	340	120 225	0.2
	PS	Polystyrene (Heat resistant)	8000	3 40	0.1	10 350	GP G	870	221	194 200	0.2
		Polyurethane Elastomer	5000	700 200	0.77	75 327	G FG		284		1.2
	PVB	Polyvinyl Butyral	500	150 450				1015			
	PVC	Polyvinyl Chloride									
	"	PVC - NonPlasticized Extrusion	7000	25 50	0	5.1 20	PG G	1230	194	140 135	0.2
	"	PVC - Nonplasticized Solvent Cast	6000	3 100	0	5 30	GP G	1230	194	140 135	0.2
	"	PVC - Plasticized Extrusion	1400	100 500	0	30 400	PF E	1015	194		0.2
	"	PVC - Plasticized Solvent Cast	3000	350 400	0	40 600	PF G	1015	194		0.2
	PSU	Sulphone Polymers	8400	20 150	2.1	18	P G	1800	374	345 358	2.6
	PVC-CP	PVC-Acetate Copolymer (Non Plstczd)	5500	3 100	0	4 20	PG G	1230	194	140 135	
	PVC-CP	PVC-Acetate Copolymer (Plasticized)	2500	100 500	0	8 150	PG G	1015	194		
Saran	PVDC-CP	Polyvinylidene Chloride Copolymer	8000	30 80		0.6 6.9	GF G	870	330	130	
	SAN	Styrene-acrylonitrile	9920	2 3	0.25			870	248	214 220	0.4
		Vinyl Nitrile Rubber	2500	250 500	0	12.5 65	G G	1408			

Notes: 1) Water absorption in % per day; transmission in cm³-mil/(100 in²-days).
2) Oxygen permeability in cm³-mil/(100 in²-days-atm)
3) "OS" refers to organic solvents.

The materials are arranged alphabetically by chemical name, which appears in the third column. The first and second columns contain selected brand names and shorthand expressions. The fourth column contains a representative film tensile strength in pounds per square inch of cross section, defined according to American Society for Testing Materials (ASTM) method D882 in almost all cases (for two or three materials, data were used from method D638, which is for bulk materials). In the fifth and sixth columns, the "elongation at break" (also from D638) is used to indicate "stretchiness". (Generally, the tensile strength is the maximum stress observed on a stress-strain curve before the material begins to rupture or stretch uncontrollably. Elongation indicates the percentage strain associated with this maximum stress.)

The next five columns give selected chemical characteristics. First, in column seven, the water absorption in 24 hours by a 1/8 inch thick specimen is indicated (ASTM D570) as the percentage increase in weight. Column eight shows water vapor transmission in the extraordinary units of gram-mils per 100 square inches in 24 hours. The next column shows oxygen permeability in analogous units. Columns ten and eleven indicate on a four-valued scale (Poor, Fair, Good, Excellent) the resistance to chemical breakdown as a result of exposure to organic solvents (OS) and water (H₂O).

Thermal characteristics follow: Thermal conductivity (in Btu-mils/(hr-°F-ft²)) and melting temperature (in °F) are self explanatory, while columns fourteen and fifteen give the temperatures at which substantial deflection will occur when the sample is subject to shear stresses of 264 and 66 psi, respectively. Finally, representative prices for some materials are included in the last column, in cents per one hundred square feet at one mil thickness.

Other information, such as area factors, detailed references and additional data gathered by Dr. A. Ravve from manufacturers and references 1-6 through 1-8, is included in the complete data base and may be obtained from the authors in printed or diskette format. This additional information includes general comments and independent values for the tensile strength and water absorption, the rated maximum service temperature, and a value for the distortion temperature at 240 psi. These data vary from Table 1-1 to some extent (20-40%); this is not a sign of error, but of the differences which can occur between two manufacturers' products, different batches by the same manufacturer, or the use of slightly different test procedures or conditions. Since no one test is directly applicable to the conditions that will obtain in chillers, these variations indicate true variations in performance that may occur in practice, and thus imply a bare minimum safety factor which must be applied in design work.

The above data were entered into a tabular spreadsheet (Lotus 123), facilitating the entry, editing and re-definition of data as the database grew. The spreadsheet also allowed flexibility in further data manipulation, which was exploited in the film selection process to be described below.

1.3 Polymer Film Selection Process

Potential applications for polymer films within absorption chillers are heat exchangers of various types, with pressure differences ranging from one tenth to two or three atmospheres between the two fluids. Heat exchange and pressure place competing constraints on the materials: the thermal conductance

Figure 1-1. Polymer tube geometry in cross section, showing static pressure (P), linear stress (S), tube diameter (D) and material thickness (t).

of the film should be as high as possible, to allow the construction of as small a heat exchanger as possible, and yet the material must be thick enough to have the strength to withstand the stress imposed by the pressure difference. Satisfying these two criteria simultaneously is the basis of our primary selection process.

For any particular design, say an air mattress configuration of tubes with a specified diameter d , the material must be able to withstand a particular maximum linear stress, that is, a maximum stress per unit length of the tube. Referring to Fig. 1-1, this maximum linear stress will be equal to the design point pressure difference P multiplied by the tube diameter D , then multiplied by an appropriate safety factor (taken as 1.0 for screening purposes); call this linear stress $S = P \cdot D / 2$. If Y is the yield or tensile strength and t the thickness of the film,

$$S = P \cdot D / 2 = Y \cdot t \quad 1-1$$

If H is the desired rate of heat transfer for the heat exchanger, k the thermal conductivity of the film, A the heat transfer area and dT the temperature difference seen by the polymer film (exclusive of fluid film temperature drops), then

$$H = dT \cdot A \cdot k / t = dT \cdot A \cdot k / (P \cdot D / 2Y), \quad 1-2$$

so

$$A = (H \cdot P \cdot D / 2 dT) / (k \cdot Y). \quad 1-3$$

H , P and dT are set by the application. There is then an engineering trade-off between tube diameter (D) and heat exchanger area (A) which also involves changes in thickness, t . Whatever the result of this tradeoff, Eq. 1-3 shows that with respect to polymer properties, heat exchanger area and, roughly, system cost, will be minimized when $k \cdot Y$ is as large as possible. Note that system cost will be minimized not only because of low heat exchanger area but also by smaller containers and piping requirements if A is kept as small as possible. Since polymer prices are volatile, we did not include it as an selection parameter at this time, but used it subsequently in design development. A similar argument based on minimizing polymer volume ($A \cdot t$) prescribes ordering the polymers on the basis of $(k \cdot Y^2)$, which leads to the same set of candidate materials when the other constraints and tests to be described are considered.

On this basis we printed out the data base with the rows arranged in order of decreasing $k \cdot Y$, the product of tensile strength and thermal conductivity. The results are presented in Table 1-2, with the bottom end of the list excluded. The quality factor, "Q", gives the product of tensile strength and thermal conductivity divided by one million. We have also eliminated from this table any candidates that did not have either good (G) or excellent (E) chemical resistance to water, and the cellulosic polymers, which have very large water absorption and transmission coefficients.

At this point we have not dealt with either the availability of the materials in film form or the dependence of their properties on temperature, a major factor for most polymers. Although temperatures range up to 300°F in an absorption chiller (excluding the first effect generator), we thought that some low temperature applications might also surface and so did not exclude any candidates from general consideration on the basis of their service temperatures. We determined availability by our ability to obtain samples rather than on vendor's assertions; success is indicated by the presence of a polymer in Table 1-3, discussed in the next section. The Acetals and the ABS were only available over ten mils thick and so were excluded from further consideration. The remainder of the samples were subjected to the tensile strength tests described in the next section.

Table 1-2
Polymer Film Candidates Ordered by
Q = the Product of Tensile Strength and Thermal Conductivity

Brand Name	Acronym	Chemical Name	Tensile Strength psi	-O ₂ ¹ Per- mea- bility	Resis- tance OS ² H ₂ O	Thermal Cond. Btu-mil Hr-F-Ft	Deflection Temp(°F)at 264 66	Q/10 ⁶
		Polyimide (Thermoplastic)	25000	25	G	G	2460 530	61.5
	PET	Polyethylene terephthalate	20000	6	G	G	1000 70	20.0
	PEEK	Polyetheretherketone	13200			E	1420 320	18.7
Nylon		Polyamide	7000	92	G	G	2320 130 300	16.2
		Acetal Homopolymer	9500				1600 255 328	15.2
	PSU	Sulphone Polymers	8400		P	G	1800 345 358	15.1
		Acetal Copolymer	8800		G	G	1600 185 311	14.1
Victrex	PES	Polyethersulphone	12200		GF	GE	1100 394	13.4
Acrylic	PMMA	Polymethyl methacrylate-standard	8200			G	1450 165 175	11.9
Tefzel	ETFE	Ethylene tetrafluoroethylene	7000	100	G	G	1650 160 220	11.6
Tedlar	PVF	Polyvinyl Fluoride	7000	3	G	G	1650	11.6
Lexan,...		Polycarbonate	8400	300	GP	G	1360 280	11.4
	PE-UHMW	Polyethylene (Ultra Hi Mol Wt)	3000		G	G	1650 110 155	5.0
	PE-HMW	Polyethylene (Hi Mol Wt)	2800				1650	4.6
	ABS	Acrylonitrile-butadiene-styrene Ionomer	5000	70	FP	G	1810 170 170	9.1
			5000	400	G	G	1780 93 113	8.9
Halar	ECTFE	Ethylene Chlorotrifluoroethylene	8000	25	G	G	1100 170 240	8.8
	SAN	Styrene-acrylonitrile	9920				870 214 220	8.6
	PVC	PVC - Nonplasticized Extrusion	7000	20	PG	G	1230 140 135	8.6
	PE-HD	Polyethylene (High Density)	2400	185	G	G	3340	8.0
Barex		Acrylonitrile methyl acrylate	9000	0.8	G	G	0 165	0.0
Korad Ac		Polymethyl methacrylate-Type A,D	5100		PG	G	0	0.0
	PVC	PVC - Nonplasticized Solvent Cast	6000	30	GP	G	1230 140 135	7.4
Aclar	PCTFE	Polychloro-trifluoroethylene	5000	15			1450	7.3
Teflon	PFA	Perfluoroalkoxy	4000		G	G	1750	7.0
Ultem		Polyetherimide	15200		G	GE	460 383 398	7.0
	PS	Polystyrene (Heat resistant)	8000	350	GP	G	870 194 200	7.0
Saran	PVDC-COP	Polyvinylidene Chloride Copolymer	8000	6.9	GF	G	870 130	7.0
	PVC-COP	PVC - Acetate Nonplasticized Copol.	5500	20	PG	G	1230 140 135	6.8
	PP	Polypropylene - Oriented	7500	160	G	G	810 120 225	6.1
	PE-MD	Polyethylene (Medium Density)	2000	535	F	G	2320	4.6
Kynar	PVDF	Polyvinylidene Fluoride	5500	14	G	G	800 188 280	4.4
Teflon	FEP	Fluorinated Ethylene-Propylene	2500	750	G	G	1750	4.4

Notes: 1) Oxygen permeability in cm³-mil/(100 in²-days).
2) "OS" refers to organic solvents.

1.4 Polymer Film Stress-Strain Data

Available data did not give any clear indication of the performance of most of these materials at higher temperatures. More importantly, all available data referred to intrinsic material strength. Since polymer heat exchangers will probably be constructed by welding films together, the strength of the welds is a matter of primary concern, and in general one would expect the welds to be weaker than the parent material due to thinning, poor annealing after heating and uneven delivery of stress if the weld is not perfectly uniform. We therefore devised a series of experiments to test the strength of the welds under conditions which might be expected in chiller heat exchangers. The samples obtained from various vendors are listed in Table 1-3 along with experimental results to be discussed below.

Polymers are visco-elastic materials which respond to applied stresses over long periods of time. This means that engineering design cannot proceed on the basis of the results of short term stress-strain testing, but must be based on the results of long term creep tests. However, it is acceptable to eliminate candidates on the basis of short term testing, since those that perform inadequately in the short term are clearly also unacceptable in the long term. Accordingly, we set up the short-term burst test apparatus shown in Fig. 1-2.

The sample is a welded cylinder of a film of the polymer being tested. Welding the samples consists of applying a pulse of heat of specified maximum temperature and duration to both sides of the pair of films to be joined as they are squeezed together under substantial pressure. In our lab this is done with a commercial film welding machine (Vertrod 14B/BHP-WC Thermal Impulse Heat Sealer). Since different polymers require different temperatures and durations for optimal welding, these parameters were determined in preliminary testing based on "T" peel tests of one inch wide strips of film welded together. These peel tests were carried out at room temperature using an

Figure 1-2. Burst test apparatus.

hand-held force gauge which retained the maximum applied force during a steady pull. The weld parameters which provided the strongest welds by this definition were used for all subsequent fabrication and testing for that material. The resulting nominal weld temperatures for our machine are indicated in the fourth column of Table 1-3; they vary with thickness for a given material. For these thin films we established that there was never an advantage to increasing the duration of the heat pulse above the minimum time period associated with the thermal inertia of the apparatus. It was at this point that the most promising material from Table 2-1, thermoplastic polyimide, was eliminated. Although the pure film has extraordinary mechanical properties, it proved impossible to make strong, reliable welds. (There are no adequate adhesives for this or other polymers, in our experience.)

Tubes two inches in diameter were slipped over the teflon end pieces shown in Fig. 1-2 and fastened to "O" ring seals with compression clamps. The tube was then immersed in a bath of fresh water at the desired temperature, filled with water and allowed to come to thermal equilibrium. The constant volume pump was turned on at a fixed rate and the cylinder would expand, resulting in a strain in the polymer sample which increased linearly in time and an accompanying increase in hydrostatic pressure inside the tube which produced a proportional stress in the material. By recording the pressure on a strip chart recorder we were thus able to generate stress-strain curves for the polymer under test; an example is shown in Fig. 1-3.

Figure 1-3 Stress-strain curve from burst tests of PET (3 mil Type A Mylar; squares) and PEEK (2 mil amorphous Talpa 2000; "+")

Table 1-3
Burst Test Samples and Test Results

Acronym	Brand Name	<----Low Temp Burst Test----->									<-----Hi Temp Burst Test----->						
		Weld Temp (F)	---Time---			Max Gauge Press (Psi)	Max Int. Stress (Psi)	Bath Temp (F)	---Time---			Max Gauge Press (Psi)	Max Int. Stress (Psi)	Slctd for Creep Test			
			Thkns (mils)	Bath Temp (F)	Zero to (sec)				Fail Mode (1)	Zero to (sec)	Fail Mode (1)						
PET	Rynite Mylar Type D	3	300	100	NT					201	333	135	WC	3.25	1083		
"	Rynite Mylar Type A	3	225	100	NT					197	14	4	WC	7.25	2416	*	
"	BCF (amorphous)	3	300	106	15	3	MP	14	4667	200	-	-	S	0	0		
PEEK	Talpa 2000(amorph.)	1	200	100	NT					201	60	65	WP	3.5	3500	*	
"	" (cryst.)	1	400	100	NT					201	18	0	WC	5.5	5500	*	
"	" (cryst.)	2	500	100	NT					201	27	0	WC	14.5	7250	*	
PES	Talpa 1000	1	500	100	NT					201	0	0	WC	0	0		
"	Talpa 1000	2	450	100	NT					201	11	11	WC	3.9	1950	*	
PMMA	Acrylic	3	150	101	27	51	WC	2.0	667	200	NT						
PVF	Tedlar (400SG20SE)	4		109	22	20	WP	12.8	3200	201	24	129	WP	3.5	875		
"	" (TWH10SS3)	1.2	400	100	6	42	WC	2.1	1750	200	0	0	WC	0.5	417		
"	"	1.2	350	101	10	78	WP	2.8	2333	200	NT						
ETFE	Tefzel	5	350	100	90	0	WC	15.5	3100	201	90	210	S	7.5	1500	*	
PE-UHMW	HD960	3	150	100	42	90	WP	7.8	2600	200	NT						
"	HD970	5	150	100	45	300	S,MP	11.0	2200	200	NT						
PE-HMW		3	250	101	30	0	WP	8.0	2667	200	60	360	S,MP	2.5	833		
PE-HD	P-9	5	260	101	42	196	S,WP	24.7	4940	200	NT						
"	HD955	3	150	100	36	165	WP	7.5	2500	200	NT						
"	HD955	3	125	100	60	200	WP	7.8	2600	200	NT						
"	GM9255F2	3	200	102	42	90	WP	20.5	6833	200	NT						
ECTFE	Halar	4	375	65	44	0	WC	18.5	4625	200	105	900	SS,MC	5.25	1312	*	
"	"	4	375	65	22	0	WC	18.4	4600	200	NT						
"	"	4	375	66	35	0	WC	18.8	4700	200	NT						
PCTFE	Aclar	5.5	Broke on assembly - too brittle														
PFA	Teflon	5	450	101	180	250	S,MC	10.9	2180	200	690	420	S,MC	7	1400	*	
PEI	Kemid	2	250	100	9	0	WC	6.5	3250	200	21	0	WC	7.7	3850		
PS	(Heat resistant)	2.5	Broke on assembly - too brittle														
PVDC-COP	Saranex 13	3.5	150	101	45	195	S	3.4	971	200	0	0	WC	0	0		
"	Saranex 15	4	150	101	102	270	S	3.8	938	200	0	0	WC	0	0		
FEP	Teflon	5	350	101	18	0	WC	7.9	1580	200	0.6	0	WC	1.75	350		

1) Failure Modes: W - on weld; M - in material; P - Persistent pinhole; S - persistent Stretch; C - Crack
2) NT = Not tested (sample availability or anticipated failure).

Here are the details: the standard rate for the pump was $R = 0.980 \text{ cm}^3/\text{sec}$. The volume of the cylinder is given by $V = \pi D^2 L / 4 = C^2 L / (4 \pi)$, (initially 220 cm^3), where C is the circumference of the tube. To the extent that the tube remains cylindrical, $(dC/dt)/C = R/(2V)$ and if the paper moves through the recorder at k in/sec, $dC/C = R/(2V*k)$. Thus for short times, when V is still close to V_0 , the strain is directly proportional to distance traveled along the chart paper, and for larger V (corresponding to stretchy polymers) it can be readily calculated. However, in most cases where substantial stretching occurred, the sample lost its cylindrical shape, compromising the geometrical assumptions behind these calculations. The nominal strain at maximum stress and the strain when stress had returned to zero (and the sample totally ruptured) may thus be computed from the time to maximum stress and the time back to zero stress if desired; since only the time data are completely rigorous they are all we have included in Table 1-3.

The hydrostatic pressure, which is measured directly by a transducer and recorded on the chart paper, is shown in Fig. 1-1 to be proportional to S , the linear stress on the sample. The internal stress in the polymer is then given by $S' = S/T$, where T is the thickness of the sample, so $S' = P*D/(2*T)$ and could be read directly off the chart paper. Fig. 1-3 is shown already calibrated in terms of internal stress in the polymer film.

The data for the samples tested is shown in Table 1-3. Two samples proved too brittle to be mounted for the tests and were eliminated; some samples (the polyethylenes, for example) tested adequately at low temperatures but collapsed totally at 100°C so that data could not be taken for them. A few tests were limited by sample availability problems, leading to the remaining blank spots in the table. On the basis of these results, those materials marked by an asterisk were selected for the final round, high temperature creep tests.

1.5 Polymer Film Creep Data

As discussed at the beginning of this chapter, stress-strain data is not an adequate basis for polymer engineering due to the phenomenon of long term creep. Creep refers to a slow, permanent deformation of the polymer in response to steady, moderate stress. It is common to all polymers (and to metals at elevated temperatures). Design of systems must include steps to assure that this deformation will not compromise the structural integrity or functionality of each component.

Creep data is available (1-1) for some materials, but only at lower temperatures, and never with references to welded thin films. To be sure that our candidate materials would be able to withstand significant pressures over periods of months and years we subjected the leading candidates to appropriate stress in a bath of 220°F lithium bromide brine for a period of one year while observing the resulting strain. Samples of polysulphone (PSU) arrived too late to be included in the burst tests reported in Table 1-3, but with such good manufacturer's specifications that they were included in these creep tests. Several of the candidates survived the test with zero or predictable and stabilized creep. In particular, all PEEK samples and PFA Teflon appear to be completely acceptable at up to 220°F and Halar may be marginally acceptable. PES, PSU and PET all appear to be unacceptable in the same range, although they may tolerate conditions at lower temperatures if suitable

applications are found. PEEK may even be acceptable at higher temperatures.

The apparatus is shown in Fig. 1-4. One inch wide strips were welded into loops and stretched on the apparatus as shown; they were then lowered into the hot brine and if they did not immediately break or stretch, were left for up to one year. The apparatus was weighed after being immersed in the brine, to compensate for buoyancy effects, and the nominal internal stress in the sample could then be calculated from the weight and the width and thickness of the strip. The tank was covered to inhibit evaporation, but no attempt was made to exclude oxygen and the brine was stirred continuously. The stirring motor was mounted on the same frame as the sample holders, leaving the samples subjected to continuous vibration as well as steady tension.

Figure 1-4. Creep test apparatus; it is immersed in hot, stirred brine up to the indicating scale. Made from 1/8 inch stainless steel rod and brass weight.

Several of the samples selected for creep tests were discarded because they stretched or broke either immediately upon entering the brine or after only a few days submersion. These "failures" are listed in Table 1-4, along with their failure modes. The "successes" are also listed in Table 1-4, with the observed creep (presented as strain, the fractional increase in length). The creep data is presented as a function of time in Fig. 1-5. PEEK was clearly perfect, with no observable creep at any time. The polysulphone performed well at first, then failed catastrophically. The PFA Teflon is also satisfactory with respect to creep, calling into question the 166°F deflection temperature reported in Table 1-1. We have double checked in Ref. 1-1 up through the 1988 edition and the datum remains, but it may well be an

erroneous reference to °C (which would be 330°F). The Halar also performed adequately, although the observed extension may have contributed to an apparent weakening indicated in the pull tests (below). The samples were carefully weighed and measured before immersion and after immersion and drying; no mass loss (or gain) was observable.

Table 1-4
One Year 220°F (105°C) Immersion Samples and Test Results

Sample Name Number	Manufacturer	Thick- ness (mils)	Weld Temp (C)	Creep Stress (Psi)	Max Creep (%)*	Pull ---Stress--- (Psi) (vs. New)
1 PEEK(c)	Talpa 2000	Mitsui	1.9	500	2260	0 5160 71%
2 PES	Talpa 1000	Mitsui	2.1	400	2000	F1
3 ECTFE	Halar	Allied Chem.	4.3	375	1000	6.9 510 39%
4 PFA	Teflon	Dupont	5.3	450	810	1.5 1870 133%
5 PVF	Tedlar	Dupont	4.0	350	1180	F1
6 PEEK(c)	Talpa 2000	Mitsui	1.2	400	3580	0 7750 141%
7 PES	Talpa 1000	Mitsui	2.3	400	1870	F1
9 PSU	Thermalux	Westlake	3.4	450	1260	F2 1050 NA
10 ETFE	Tefzel	Dupont	5.0	350	860	F1
11 ETFE	Tefzel	Dupont	5.0	350	860	F2
12 PES	Stabar S100	ICI	2.7	400	1560	F1
13 PET	Mylar	Dupont	3.0	225	1580	F1
14 PET		BCF	3.0	300	1430	F4
15-17 PEEK(a)	Stabar K	ICI	2.0	450	2150	F1
18 PEEK(c)	Stabar XK300	ICI	2.0	550	2150	0 7400 103%

*F1: Broke within one hour; F2: Broke after 30 days;
F3: Broke after 300 days; F4: Stretched within one hour.

The surviving samples were also used in pull tests to compare the strength of the welded joints after one year of submersion to that of virgin joints tested earlier. The pull tests were carried out using the hand-held force gauge testing 0.5 inch-wide strips of immersed sample including the weld joint. In all cases failure occurred at the edge of the weld. Two different strips were tested from each sample, and the maximum pull stress from the two samples agreed to within 20% in all cases and within 10% in most. The average pull stresses are indicated in Table 1-4 and compared to the peak stress of the burst tests from Table 1-3. The scatter in the results for PEEK indicates that there is in all probability no deterioration at all. Our single Halar sample may have become somewhat weakened.

Infrared absorption spectra were taken of all samples and compared to spectra from virgin material. With one possible exception, the results indicated no discernible change in chemical structure. The spectra were taken from 600 to 4000 cm⁻¹ (wavelength from 2.5 to 17 microns), and both virgin and immersed samples were cleaned with ethanol and distilled water to remove contaminants. In these spectra, various clearly delineated absorption lines can be associated with the chemical bonds holding the polymer together. A

change in the spectrum, such as the appearance or disappearance of a line, or even strong weakening or growth, can indicate that a particular chemical bond has been attacked by the environment. No such changes were observed for any of the samples with the exception of PEEK samples 1 and 18, where a modest absorption line, present in the virgin samples between 2570 and 2600 cm^{-1} has disappeared in the immersed samples. This could indicate a breaking bond in the PEEK, but we were not able to identify the original absorption line with any lines expected on the basis of PEEK's chemical structure, and the line did not appear at all in the third PEEK sample (number 6). We therefore believe the line in the virgin samples to have been caused by some residual lubricant or other processing contaminant which leached out during submersion.

Figure 1-5. 212°F creep test results for PEEK, Halar, PFA and polysulphone.

The physical condition of the immersed samples appeared unchanged with respect to color, ductility, surface striations and gloss with a few exceptions: all samples had taken a permanent set where they had been bent around the support rods under stress. This was less apparent in thin samples and more so in thick samples, and the PEEK and PFA Teflon remained ductile. The Halar was ductile, but had gone further in setting, in that it had molded itself to irregularities in the rod surface and now contains a permanent image of those irregularities. The polysulphone had hardened and become quite brittle at the set point, consistent with its early failure.

All samples had a tan, friable deposit on them which had not fallen off despite the continuous shaking and their vertical orientation. The deposit was up to 0.5 mm thick and would clearly have had a devastating effect on heat

transfer. Since this was an open, aerated tank the deposit was almost certainly corrosion products from the various metals present (the deposit was also present on the bottom of the tank), and was often streaked with green, indicating copper oxide leached from the brass weights. Since a chiller would be a closed system with many orders of magnitude fewer corrosion products, we do not regard this deposit as a problem, but note that this experience demonstrates that the polymer films are not so fouling-averse as to be self-cleaning.

Some potential applications call for heat exchange materials operating at 300°F; it would be foolhardy to assume that successful performance at 200°F indicates anything about 300°F performance, and time and resources precluded another full set of year-long creep tests at 300°F. We did, however, run short term tests on various crystalline Mitsui PEEK samples in a convection oven at 300°F. The support rigs of Figure 1-3 were used again, but this time there was no buoyancy force due to brine and the total force on the samples was 5.2 lb. This gave 2.2 kPsi internal stress for a one inch wide, 2.3 mil thick sample, 2.7 kPsi for 1.9 mil samples and 4.2 kPsi for a 1.2 mil sample. For the thicker samples, we found an initial 3 to 6% strain in the first hour, with modest additional creep thereafter; the 1.2 mil sample was clearly not strong enough. This is different from the 212°F performance, where there was no creep at all, but it is a level of performance that can be used in design work if it holds true over longer time periods, since it indicates that slightly thicker film might not creep at all; clearly more testing is needed. The results of these tests are plotted in Fig. 1-6.

Figure 1-6. 300°F creep tests of PEEK, showing internal stress in kPsi.

1.6 Final Polymer Film Selections

Drawing on the large amount of data accumulated, we now make specific recommendations for the use of polymers in the heat exchangers comprising an absorption chiller. Referring to Figure 0-1 of the Introduction, showing the components of a directly fired double effect absorption chiller, we note that in the first effect generator the heat exchanger is exposed to gas flames. Even if these flames are confined to the surface of a catalytic burner the environment is too extreme even for PEEK. In a steam fired unit it would be equally difficult to make use of polymers, since the steam would be at over 300 °F and 70 psi, placing the thin films under unacceptably high pressures. Accordingly, we drop the first effect generator from consideration.

The absorber and condenser run at low temperatures and therefore would appear to be prime candidates for a variety of polymers. However, they are both in contact with water which runs through the wet cooling tower and which is therefore thoroughly aerated. A glance at the permeability ratings in Tables 1-1 and 1-2 indicates that large quantities of oxygen (and other gases) would quickly penetrate to the interior of the chiller. In addition to inhibiting heat and mass transport as non-condensables, the oxygen would rapidly promote corrosion of any parts of the chiller which were not made of polymers, including the first effect generator and a certain amount of the structure and piping. If the corrosion problem could be minimized, the non-condensables would still have to be removed through more frequent use of the pumping techniques used in today's chillers, at a substantial operational cost. Until a solution to these problems can be found, we will drop the absorber and condenser from consideration.

The evaporator poses similar problems. The permeability of the polymers ensures that system water and liquids or gases dissolved in it will diffuse into the chiller over time. The water itself is not a problem, since it can simply be pumped out of the evaporator sump from time to time. If the building distribution system is itself sealed, the chiller will act as a de-aerator, pulling oxygen and nitrogen out of the system water until they are depleted; this cycle would be repeated whenever make-up water was added. The problem could be ameliorated by using de-aerated system water, but since we are concerned with very small quantities of oxygen when preventing the corrosion of thin metal foils, this would be an extremely demanding procedure and would make the chiller unattractive in many applications. We would also be demanding that the building distribution system itself be vacuum tight, a much more stringent requirement than is common in practice, and one whose cost would have to be written off against the savings in chiller construction. Faced with these problems, we have decided to omit the evaporator from further consideration as a candidate for polymer film construction at this time.

This leaves the three internal heat exchangers as candidates, since permeability to water and gases is not an issue inside the chiller, and we consider them in turn. The second effect generator operates under a differential pressure of 0.85 to 1.0 atmospheres, with pure steam condensing on one side and dense brine boiling on the other, at a temperature of up to 210°F and a temperature differential of about 15°F, with a heat flux of 2500-3000 Btu/hr-ft² (1-6). Since the tests reported in Table 1-4 were based on a one inch wide sample carrying a load of 4.3 lb., consideration of Fig. 1-1 will show that tubes of diameter 0.6 inches under a differential pressure of one atmosphere will subject the polymer films to the same internal stress as

obtained in the tests. Metallic heat exchangers have essentially zero temperature drop in the material. Will polymer films of these thicknesses transfer adequate amounts of heat? Combining the heat transfer coefficients of Table 1-1 with the thicknesses of Table 1-4 and demanding a heat flux of 2500 Btu/hr-ft², we find temperature drops in the polymer film of 2.1 and 3.5°F for the 1.2 and 2 mil PEEK and 7.1°F for the 5 mil PFA Teflon. As discussed below, these should be small compared to the temperature drops in the static films of liquid next to the heat exchanger surface, about 20°F for these heat exchangers.

Thus PEEK incurs completely acceptable thermal resistance, and PFA may be acceptable. If Halar proves mechanically satisfactory after all, this sample would still give rise to a temperature drop of 10°F, which is almost certainly too high. Halar might be acceptable, and PFA more acceptable, in a design with smaller tube diameters, which would permit the use of proportionally thinner films. Here, however, the fraction of material used up in welds increases, lowering economic performance; more detailed design work needs to be done before a decision is made on this issue. We conclude that certainly PEEK, and perhaps PFA Teflon, are acceptable candidates for a second effect generator if they are welded into tubes with diameters of 0.6 inches or less or used in other designs where they will be subjected to internal stresses comparable to or less than those in the creep tests. Such a design will be discussed in Chapter 11.

In our Phase I Report (1-7) we determined breakeven materials costs of \$1.40/ft². Table 1-1 indicates that our 1.9 mil PEEK will have an approximate resin cost of \$0.20/ft², the 5 mil PFA will correspondingly cost \$1.10/ft² and the Halar, \$0.45/ft². (We made a small-lot, special order of 100 ft² of PEEK at \$2.50/ft².) At this point the PEEK appears to be quite advantageous economically, and it appears that our cost goals will be met easily.

The low temperature recuperator operates in counterflow with the cool stream at a pressure a little over one atmosphere above the hot stream. The maximum temperature in the device is about 200°F and the temperature drop between streams is between 13 and 30°F. Current designs carry 1150 Btu/hr-ft². (Ref. 1-9, section 3). The arguments in the preceding paragraph show that PEEK and PFA films would serve more easily in this application than in the second effect generator. The use of narrower tubes, less of a design problem in this liquid-to-liquid heat exchanger, might also make Halar a more likely potential candidate. The breakeven materials cost goal of \$0.90/ft² is sufficiently low that PFA might prove too expensive, but both PEEK and Halar seem capable of meeting the goals easily.

The high temperature recuperator operates under more stringent temperature requirements, up to 305°F, again with a maximum pressure differential of a little over one atmosphere. Power densities are 1850 Btu/ft². If the materials can take the higher temperatures without creep the heat transfer characteristics appear to be adequate. The maximum service temperature for PEEK and PFA is 480°F, which is acceptable but gives no indication of creep. The short term creep tests discussed above indicate that PEEK may be a viable candidate for the high temperature recuperator as well. Tests were not run for PFA, largely because its high cost indicates that PEEK would be a preferable material even if PFA performed adequately. A set of long term creep tests in brine for both candidates at 300°F would be a highly desirable next step in any future work in this area.

Finally, an entirely different approach to the manufacture of heat exchangers from polymer films was suggested by A. Ravve: use inexpensive polyethylene, strengthened by cross-linking induced by electron beam irradiation, impregnated with graphite fillers for additional strength and enhanced thermal conductivity. We think this is an exciting possibility, but were unable to find ready-made impregnated films. Unfortunately, the expense of having film rolled to order, and the additional high costs for small-lot irradiation precluded any experimental work on this idea in this phase of our work. Dr. Ravve's proposal is included as an appendix.

2. POLYMER HEAT EXCHANGER FABRICATION

2.1 Introduction

In Chapter 1 we identified three absorption chiller heat exchangers which were likely candidates to be built from a polymer film: the second effect generator and the two recuperators. In this chapter we discuss the design, construction and testing of one of these, a miniature recuperator made from PEEK. The recuperator, for simplicity in construction and in keeping with one of the overall design options proposed in Chapter 11, involves the counterflow of two fluids, the cool fluid being under higher pressure and confined to a set of tubes while the hotter fluid flows more freely around it.

Performance of the heat exchanger was analyzed with Wilson plots and found to be typical of a counterflow recuperator. Because time and design constraints restricted us to a design involving quite low fluid flows and high film resistance, we could not observe any effect of the moderate thermal conductivity of the polymer. The low effectiveness indicates the importance of higher fluid velocities. We have not identified any problems which would prevent the adaptation of techniques used in metallic heat exchangers to ensure this condition. We have therefore demonstrated that a polymer film recuperator can work at moderate pressures and fluid velocities and have designed a manifolding technique using blow molding which will allow substantial improvements in performance in a succeeding generation of these devices.

2.2 Materials Selection

As discussed in Section 1-7, PEEK appears to be the first choice for a recuperator material, largely because on a unit mass basis it outperforms PFA Teflon by a substantial margin. This allows us to cut the cost of a recuperator significantly by using less material. It also allows the recuperator to work more effectively because the use of thinner material leads to lower temperature drops across the polymer film. Using the data of Tables 3-5 and 3-6 of Ref. 1-9, for example, there is a heat flux of 1150 Btu/hr-ft² in the low temperature recuperator. Using the thermal conductivities of Table 1-1, we find that the 1.2 mil PEEK would incur a temperature drop of 1.5°F (0.8°C) while the 5.3 mil PFA Teflon would have a corresponding drop of 8°F (4.5°C) across the material. Since the approach temperatures are approximately 15 to 20 °F (8 to 11°C) in this recuperator, use of the PFA would produce a significant change in the operating parameters, while use of PEEK would not.

PEEK is also a reasonable choice because it may prove viable at 300°F (150°C), the highest operating temperature in the high temperature recuperator, while PFA Teflon almost certainly will not. This would allow both recuperators to be made to the same specifications, a significant advantage in system cost and complexity. PEEK's ready availability and ease of welding are two more factors in its favor, although PFA Teflon is comparable with respect to these characteristics.

Figure 2-1. PEEK shell and tube recuperator; dashed lines are welds, clamps hold polymer film to manifold tubes.

(a) Section A-A'

(b) Section B-B'

Figure 2-2. PEEK shell and tube recuperator, sections from Fig. 2-1.

2.3 Design and Fabrication

The goal of this task was to demonstrate the operation of an all-polymer heat exchanger; we have done this, but because of constraints of time and resources, it was not possible to produce a recuperator which would be considered sophisticated by today's standards, and as a result performance, while quite adequate for our experimental purposes, was not outstanding. The reasons for this are discussed below, as are the numerous steps which can be taken to enhance heat transfer and increase the effectiveness of the recuperators.

First it was necessary to settle on a basic design. In our prototype chiller design, one of two options for the recuperators consists of coiled spiraling tubesheets in the bases of the annular spaces comprising the first and second effect generators (see Figure 8-2 of Phase I Final Report). The tubesheets will contain the lower temperature weak solution under about one atmosphere pressure, and will be immersed in the hot, low pressure strong and intermediate solution flowing off the inner surfaces of the two generators. As a simplified prototype for this, the recuperator built in this task is a simple sheet of tubes containing cool fluid resting in a bag (or shell) carrying a (relatively slowly) counterflowing hot fluid. The basic design is shown in Figures 2-1 and 2-2.

The only design constraint other than those imposed by the material characteristics was our insistence that it be possible to construct the design by conventional thermal welding, using our welder. We investigated the possibility of using locally available ultrasonic welding (a different machine from our ultrasonic metal welder), but it seems to hold no immediate advantage over our thermal welder except with respect to attaching fittings. Efficient manifold design is a significant problem, and being able to readily attach fittings a strong advantage, so that ultrasonic welding may well be the best way to fabricate many components in the long run, but for the current project the flexibility of being able to re-work the piece on the spot outweighed other considerations.

Very high levels of sophistication are common in heat exchanger design these days (2-2,2-3), in particular involving the enhancement of performance by lowering the fluid film heat transfer coefficients through surface geometry-induced irregularities in the flow paths. We considered various designs which would permit compact design and promote irregular flow such as stiff polymer inserts and folded flow paths. We are sure such techniques will play a major role in future polymer film heat exchangers, especially because the weldability of the polymer films makes them readily adaptable to a wide variety of shapes and configurations. However, the complexity of careful design and the time required for tooling compelled us to leave these techniques aside in this first-generation effort.

One particularly vexing problem inherent in our relatively primitive fabrication techniques results from pressurizing an object, such as the sheet of tubes or shell used here, after it has been fabricated from flat sheets of polymer film. High performance materials like PEEK are intrinsically rigid, and the inflation these structures underwent as a result of pressurization often resulted in crimps that would cut off liquid flow from whole sections of the tubes. This problem was dealt with in this phase through careful weld design, the use of manifolds (discussed below) and relatively low pressure

operation. In later phases, a complete solution to this problem can be worked out using high temperature blow-molding of the films to produce precisely controlled three dimensional shapes including appropriate manifolding. Although we have ovens adequate to this task, we did not have the time to prepare suitable molds or prepare the pressurizing apparatus, and this next obvious next step had to be put off for later development. The use of dissimilar films to promote specific, desired distortions of components at elevated temperatures was similarly postponed.

Thus the device we built and tested was the relatively simple structure shown in Figure 2-1. Thirty inches long, the tube assembly itself consists of 18 parallel tubes welded from a single sheet of PEEK, with input and output manifold tubes inserted into either end. The sheet of PEEK was 18 inches wide and this results in a set of tubes with diameters of 0.56 inches when the 1/8 inch width of the welds is taken into account. Since both sides of the tube assembly are wetted this results in a total heat exchange area of 3.3 ft². This entire tube assembly was then welded inside a bag which constitutes the shell for a shell and tube system. Welding the one unit inside the other while providing manifolding tubes and not accidentally welding them shut required constant attention to detail, although once organized the work is straightforward and could readily be automated. Prior to construction of this unit several prototype tube sheets were prepared from inexpensive materials to develop welding and masking techniques and to determine acceptable geometries.

The dimensions were chosen under severe time constraints; we would have much preferred to test a larger recuperator. Using the data of Tables 3-5 and 5-3 of Ref. 2-1 we estimate a liquid film heat transfer coefficient in those recuperators to be 150 Btu/hr-°F-ft², and use this number even though our more slowly moving fluids would certainly give rise to some lower value. Applying the methods of Chapter 2 of Ref. 2-2, especially Figure 2-12, we estimated that we would observe an effectiveness at 1.0 gpm of 35%, enough to allow reasonable testing.

For testing, the entire bag was placed inside a box constructed of slabs of insulating material which also provided some structural support, preventing the outer bag from blowing up like a balloon; similar support, although probably not insulated, would be provided by a structural polymer frame inside a chiller. In the design of Chapter 11, a separate bag might not be necessary if the tubes rest in the pool of fluid at the base of the chiller.

In the prototype as constructed in our lab, manifold tubes were necessary inside both the shell and tube assembly to ensure uniform distribution of the fluid and to prevent excessive warping of the polymer film objects under internal pressure, a process which tended to seal off passages if not controlled. These manifolds are indicated in Figs. 2-1 and 2-2. In this prototype work these tubes were made of copper as a matter of convenience; in later versions these rigid tubes would be made of some polymer with adequate heat tolerance, perhaps PEEK or PFA Teflon, and would be welded to the film, giving an integrated structure which would be rigid where necessary. Clearly the manifold tubes comprise a sufficiently small area that their heat exchange is not important and they can therefore be made of quite thick material.

2.4 Testing and Heat Transfer Performance

The heat exchanger test facility is described in detail in Chapter 6 in order to preserve the original numbering of tasks in this final report. Logically, it would be useful for the reader to skip ahead and read that (short) chapter now in order to understand the operation of the test facility clearly. In essence it provides the temperature and static pressure at each fluid entry and exit point and the fluid flow rate through both legs of the system. The cold water ran through the tubes for these tests, and the hot side is the shell surrounding them. Data were taken for several different flow rates through each leg of the recuperator. At each point a heat balance and the effectiveness were calculated, and the results are presented in Table 2-1. (Runs 11-16 are omitted due to measurement errors.) The relevant formulas are given in Chapter 6.

Table 2-1. Performance of PEEK Shell and Tube Recuperator

Run #	F _h (gpm)	F _c (gpm)	T _{hi} (°F)	T _{ho} (°F)	T _{ci} (°F)	T _{co} (°F)	Heat Balance (%)	Effective- ness (%)
5	.719	1.273	149	125	79	92	-4.3	.335
6	.739	1.148	149	125	77	91	-10.4	.317
7	.739	.899	151	129	76	93	-6.4	.284
8	.663	.480	151	132	75	102	2.9	.350
9	.697	1.435	152	125	74	87	-0.9	.344
10	.593	1.571	152	121	74	85	-6.5	.385
17	.518	.926	145	117	74	88	-12.0	.373
18	.846	.934	146	124	74	91	-17.3	.282
19	.971	1.385	147	124	74	88	-15.3	.294
20	1.627	1.571	149	133	74	93	12.8	.237

* Subscripts: h = hot side, c = cold side, i = input end, o = output end.

In addition, the data of runs 5 through 9 were used to generate a Wilson plot to illustrate various heat transfer properties; the plot is presented in Figure 2-3; Wilson plots are reviewed in Chapter 6. For analysis, we will assume that the average flow rate, .711 gpm, obtained for all five runs. The velocity for the cold side was calculated assuming 18 parallel tubes of 0.65 inches diameter giving a total cross section of 6.0 in², and for the hot side the cross section was taken as an 18 in² rectangle minus the 6 in² occupied by the tubes.

Then a linear regression on the five plotted points yields an intercept corresponding to a thermal conductance of 1.01 Btu/hr-°F-ft² due to the hot side film resistance and the wall material, and a slope such that the conductance due to the cold side film is given by 58*V_c^{.8}. Although these are quite low values of thermal conductivity, due to the slow motion of the water through our sub-optimal design, they clearly demonstrate that the polymer recuperator is acting just like a metal recuperator: first, the coefficient of 58 for the cold-fluid-dependent part of the conductance is to be compared to a

value of 77 for the copper tube-in-tube device tested as a base line and described in Chapter 6; given the extreme differences in construction, the agreement between these coefficients clearly indicates that engineering polymer recuperators is just like engineering metal ones, heavily and similarly dependent on managing boundary films.

Figure 2-2. Wilson plot for PEEK shell and tube recuperator.

Second, the high cold side velocity limit of $1.01 \text{ Btu/hr-}^{\circ}\text{F-ft}^2$ is just what one expects if the same slope applies to the hot side fluid, moving at a velocity corresponding to 0.7 gpm, giving $1.06 \text{ Btu/hr-}^{\circ}\text{F-ft}^2$. (In fact, this 5% agreement must be fortuitous.) The calculated conductance of the 1.9 mil PEEK film is $2700 \text{ Btu/hr-}^{\circ}\text{F-ft}^2$, far too high to be observable in this device.

We conclude that we see no barriers to constructing recuperators of PEEK (or perhaps PFA Teflon) that will have the same thermal characteristics as current metal devices, will be completely corrosion free and can be blow molded and formed in ways that are at least as suitable for automation as current techniques for metal recuperators are. This will be important, since the PEEK will not offer a cost advantage over the $\$0.75\text{-}1.00/\text{ft}^2$ materials cost of carbon steel. (2-1) The flexing and vibrating of the polymer film, and its inherent chemical inertness, may serve as anti-fouling characteristics, but this will require testing since the submersion tests reported in Chapter 1 showed that the PEEK is not completely immune from fouling if the brine is sufficiently dirty. The freedom from corrosion will mean that the initial shakedown period undergone by chillers, when the formation of oxide layers on the steel results in the emission of significant quantities of hydrogen which must be pumped out regularly, will be shortened and lessened in intensity by a significant amount, resulting in lowered installation and startup costs.

3. POLYMER ELECTROPLATING FACILITY

3.1 Introduction

Another approach to heat exchangers of low material intensity and high corrosion resistance is to use a base metal, such as carbon steel, with a corrosion resistant polymer coating to protect it from the corrosive agents. Such a coating can in principle be applied in several ways, including solution casting and electro-deposition. Heat transfer analysis indicates that the coating should be one mil thick or less, however, and this turns out to be a fairly restrictive condition when we also demand that the coating be flawless. (Clearly nicks or bubbles will allow the LiBr access to the steel, and the ensuing corrosion will lift the polymer from the rest of the steel). Most experts consulted agreed that it would be extremely difficult to obtain a flawless one mil coating by solution casting or painting techniques. Apparently tiny inhomogeneities or dirt specks on the surface prevent full coverage unless the coating is very thick. We have accordingly dropped solution casting from further consideration.

This has led us to examine electrocoating techniques. Here the sample to be coated is immersed in a solution containing the polymer in an ionized state, and a voltage established between the solution and the sample leads polymer ions to deposit on the sample. As they build up an insulating layer, current and polymer deposition is directed to the areas that are not yet coated, resulting in an extremely uniform coating.

In this task, we set up a polymer electrocoating facility and used it to test three different coatings, an anodic epoxy coating from Glidden and cathodic epoxy coatings from Glidden and PPG on aluminum, copper and various steels. The coatings were tested for integrity, durability, flexibility and other relevant properties. Their corrosion resistance was examined in Task 4 and their heat transfer characteristics in Task 5, and the results for those tests are documented in those chapters of this report.

In summary, we found that one "torture test" allowed us to choose one paint as more appropriate to thin metal foil construction. After normal inspection and testing (discussed below) the foil samples were bent into a "U" shape, squashed under a standard force of a few pounds and flattened out again. Both sides of the sample were then subjected to an electrical conductivity test of coating integrity again, to see if this treatment had produced cracking. In most cases it did for both of the Glidden paints, while it almost never did for the PPG paint. This is consistent with the appearance of the paints, for the Glidden coatings do appear harder and more brittle. Thus although the Glidden paints may well be as good or better performers on rigid substrates, the PPG coating is clearly superior on these thin, flexible foils and will be used exclusively in our work.

Two negative findings emerged: first, it seems to be impossible to deposit a complete coating around the sharp edges of foil where it has been cut, a problem which can be overcome by painting a solvent-deposited bead of sealant around the edges of coated objects. Second, we have not found any way to coat the interiors of tubes or similar objects, so we can only use this technique in components where the interior surfaces of a heat exchanger are not exposed to corrosive fluids. The design implications of this are discussed at the end of this section.

3.2 Apparatus

In this and the following sections we discuss the apparatus and procedures which were common to testing of all three coatings and the different metal substrates; specific exceptions will be noted below as necessary. The apparatus used for electroplating is quite simple and is shown schematically in Fig. 3-1 for the case of cathodic deposition; for anodic deposition the polarities are reversed. The variable DC power supply included both current limited and voltage limited operation, and either current or voltage could be recorded with a strip chart recorder during the deposition process. For small samples the opposite electrode was a sheet of stainless steel shaped to form a lining of the vertical walls of the coating tank (which was usually just a 0.50 liter beaker). These beakers were used with magnetic stirring rods and a stirring table to keep the solution agitated. Temperature control was found to be unnecessary: the recommended solution temperatures were slightly above room temperature, and were attained due to joule heat warming of the solutions.

Other equipment included, in addition to standard chemical apparatus, a pair of infrared lamps with a variable voltage control for pre-drying coated samples and a well regulated oven for curing them. A 50X binocular microscope was used for visual assessment of the samples.

Samples consisted of small rectangular pieces of metal strip, about one inch wide and three inches long, of which the bottom two inches would be coated. Since both sides were coated, this gave about four square inches of applied coating. The materials were a few mils thick, as shown in Table 1-4 below, except the Bonderite, which was only available in relatively thick samples. Pieces were cut with sharp shears or a paper cutter; under a microscope the result was edges which were flat and perpendicular to the sides, with sharp corners and occasional wisps of not-quite-detached metal clinging to them.

Figure 3-1. Polymer electroplating bath.

3.3 Coating Procedures

Every electrocoating experiment begins with a thorough cleaning of the workpiece. There are a variety of commercial and laboratory mixed cleaners, which are of varying efficacy on different metals. Our choices, based on initial recommendations and considerable testing, are specified in Table 3-1. It is best to coat a sample immediately after it has been removed from the cleaner and rinsed, since active metals such as aluminum and copper are inclined towards discoloration and the formation of oxide layers. Cleaning is an extremely important part of getting a defect-free, well-adhering coating and the simplicity of this table does not adequately represent the time and effort that were needed to determine these optimal treatments. Included in the table is a commercially pre-treated steel, Bonderite, which was tested alongside the samples cleaned in-house.

Table 3-1
Metal Substrates and Corresponding Cleaning Solutions

Material	Thickness (inches)	Cleaning/Preparation
Aluminum 3003	.0030	Acetone Wipe H_3PO_5 (10-30% 3-6min) (Alumiprep recommended)
Copper 161	.0030	H_2SO_4 (20%) with H_2O_2 (5%) for 1-5 minutes
Steel (tinned) 1010 Carbon	.0020	H_3PO_5 (15-20% 3-6min) Acetone pre-wipe optional
Steel (untinned) 1010 Carbon	.0020	H_3PO_5 (15-20% 3-6min) Acetone pre-wipe optional
Steel (Bonderite EP-2).028		Pre-cleaned and treated.

The coating stage itself consisted of setting current and voltage limits, inserting the sample in the bath and turning on the power. The circuit is essentially a simple series circuit with a single resistance which is proportional to the total charge which has flowed through the circuit (that is, to the amount of deposited polymer), and which is therefore a function of time. Initially, the resistance is small and a large current would flow were it not for the current limiting circuit in the power supply. Later, the polymer layer thickens, the resistance builds up and the voltage limit becomes the controlling factor for the last moments of the run. The current never drops to zero, as a small leakage current, less than one per cent of the maximum, continues to flow at such a low rate that no apparent deposition is taking place. At this time, deposition is essentially complete.

In our initial experiments, currents as high as 500 mA per square inch were used. At these high settings, the voltage reached the recommended final values of 175 volts after times on the order of only 30 to 50 msec. However,

these coatings were very poor, having several pockmarks or ruptures per surface. Assuming that all the joule heat is (initially) deposited in the one mil thick coating, an order of magnitude calculation shows immediately that there is sufficient energy being deposited to boil some water in the coating, undoubtedly the source of the scarring. When the current limit was lowered to 10 mA per square inch the system took several seconds to reach the limiting voltage, much less energy was deposited and the surfaces were vastly improved. This procedure was followed for all subsequent tests; specifics are given for each coating in their respective sections if the numbers vary from these.

After removal of the workpiece from the bath samples, excess paint is removed either by rinsing or by evaporation in a well ventilated area. Initial drying is carried out by a brief exposure to low level infrared heating lamps and the final cure stage is completed by a 15-20 minute stay in an ordinary oven or by leaving the sample in the infrared environment while gradually elevating the temperature. These methods were developed through considerable experimentation; for example, without the initial drying stage, water trapped in the coating would boil out during the cure, leaving a flawed surface. The pros and cons and optimal combinations of these methods are discussed in sections to follow when the three commercial paints are evaluated.

3.4 Testing of Coated Samples

A battery of tests was developed to determine ratings for each epoxy. These tests were concerned with numerous properties of the polymer coating. Included among these was the resistance to a powerful solvent and to mechanical stresses, integrity under surface scratching and the application of adhesives, and the ability to hinder electrical current which might travel through pinholes or other apertures in the coating.

Methyl ethyl ketone (MEK) was the solvent selected for the first test mentioned above. A brisk manual application consisting of sixty circular rubs with a MEK-saturated wiper was made on the coating; if the coated metal became visible, the sample was designated as a failure for the MEK test. If the solvent had little or no effect, the sample was assigned a status of passing for the test. While it might seem that a testing system with a higher degree of specificity could be used, it became clear after actual testing that samples had a pronounced tendency to either become exposed after several seconds or to remain thoroughly coated regardless of the time. The pass/fail system reflects this tendency.

The existence of tiny or even microscopic flaws in the coating was a matter of great concern, and an electrical conductivity test was developed to test for this possibility. The conductivity test was carried out by distributing lithium bromide brine over the coating surface, placing an uncoated extremity of the metal substrate in contact with an electrode, and then using a probe of opposite polarity to the attached electrode to see if and where a current could be passed. If the ammeter registered a current, minute section(s) of metal were exposed and thus the sample's ability to avoid corrosion was seriously in question. As will be discussed below, the integrity of the coating was poor at the edges of a sample, and almost all samples would fail a conductivity test at the edge, so here we report only on conductivity tests for the central portion of the flat surface of a sample. A current of a fraction of a microampere in response to several tens of applied volts was

regarded as a failure; tests of samples where visual microscopic examination revealed pinholes produced currents of several milliamperes at ten volts.

A torture test was also devised: each sample was folded and then compressed using both 19 lbs per inch of fold and then, in a different region of the sample, 8 lbs. per inch. In all cases the sample would end up folded like a piece of paper and a clear crease would form in the metal. The sample was then returned to a semblance of its original position by flattening it with the same respective weights on a hard surface. After a complete compression and stretch the resulting depressions and ridges were examined visually and then checked with the conductivity test described in the previous paragraph. Any observed current was regarded as a failure.

As a hardness test, the virgin surface was challenged with a sharp instrument to judge the ease with which penetration to the base metal could be made. Attempts were made to insure a standardized pressure each time a sample was scratched in this manner, and the following (somewhat subjective) ranking system was employed: 1-soft, 2-average, 3-above average, 4-hard.

Adhesion was tested by applying a highly viscid tape surface to the coated surface at a point where an "X" had been scratched through the coating down to the metal. The tape was then scrutinized after removal for coating which might have adhered to it. If there was no coating on the tape it merited a rating of "good"; continuous light traces of coating were called "fair", and clearly visible regions of coating received a designation of "poor". Adhesion was also tested in the bends and crevices created by the compression and stretching tests, with the results being considered in an overall adhesion rating.

3.5 Testing of PPG Cationic Epoxy

PPG Powercron 600 Cationic Epoxy coating was tested on a total of five candidate metals: aluminum, copper, sheet steel, precleaned bonderite steel and tinned steel. Of primary concern after an appropriate cleaning of each sample was a determination of the most efficient dry and cure cycle. The drying stage is intended primarily as a preparation for curing, and it is essential that all excess water and coating be removed during this stage of production. It was anticipated that the most time efficient dry cycle would be independent of the substrate to within experimental uncertainties and this expectation was confirmed. Using infrared heating lamps, it was ascertained that 40-45 °C (105-115°F) indicated on a thermometer next to the sample for a duration of two minutes was sufficient to eliminate problems such as pinholes and bubbling during the cure. This determination was made by both microscopic examination and by subjecting the sample to the conductivity test described in the previous section. The high rate of passing for this test at the above mentioned drying specifications persuaded us to adopt this dry cycle as the standard procedure.

The fairly low temperature and short duration are not due to stinginess on our part; although slightly higher temperatures and time durations achieve the same level of visual quality, freshly coated samples have a tendency to burn and dry unevenly when exposed to excessive heat (greater than 60 °C = 140°F on the thermometer) or to temperature rates of increase which exceed 3-4 °C (5-7°F) per minute when still damp from the coating bath. As became

evident after subjecting the samples to a battery of stringent tests, over-dried samples had a greater tendency to be brittle and to fail compression and stretch tests.

The cure phase of production is crucial for strengthening the polymer bonding in the coating and can be accomplished either by continuing the use of the infrared heaters at greater and greater temperatures or by heating the samples in ordinary oven. Though the quality of a successful infrared cure was comparable to a typical oven cure, the heating lamps required constant adjustment and attention to avoid burning or overheating the samples. The gradual heating in an oven, however, insured uniformity automatically and produced a surface was more aesthetically pleasing under the microscope. It is conceivable that with proper automation an infrared curing process could be perfected, but the sensitivity of a fresh Powercron 600 coating to overheating makes this a challenging task, as shown in Table 3-2. Depending on the specific requirements for a given metal, an oven cure of 200-220 °C (390-430°F) for 10-15 minutes is sufficient, while for an infrared cure one must ramp from 40 to 60-65 °C (from 105 to 145°F) over 7-9 minutes followed by a stable infrared bath at 60-65 °C (145°F) for 6-8 minutes. (Bear in mind that the infrared temperatures are thermometer temperatures measured near the open air lamps - actual sample temperatures may be somewhat different.)

After successful coating and curing, the result was a microscopically smooth, plane gray surface. On aluminum, copper and steel substrates the coating plated out to a remarkably uniform 1.1 mil thickness ($\pm$ 0.1 mil). The density of the cured coating was 1.24~~0~~.2 gm/cm³ over several samples. Mechanical properties still varied considerably depending on treatment, however, so each sample was subjected to the battery of tests and examinations after curing. The results for specific materials and cure cycles are given in the tables below. All samples passed the conductivity test on the unstressed surface.

Aluminum was employed as a substrate in an exploratory capacity while determining the most advantageous dry and cure cycle, as shown in Table 3-2. The results of Table 3-2 indicate that hotter, shorter cure periods result in greater hardness but worse adhesion, and that longer bake periods can make up for lower temperatures in resistance to the MEK test. The results of the compression/stretch tests are inconclusive but not satisfying for this set of materials. The results of these tests will depend on the substrate material: the soft aluminum will bend more sharply under a given stress than will the more resilient steel, and that may be one reason for the mediocre performance manifest in Table 3-2.

Table 3-2 also shows that oven cured Powercron 600 on an aluminum substrate performs satisfactorily in adhesion, resistance to MEK and moderate creasing if cured in the 190-220 degree range for 15 minutes; it still shows only average hardness at best.

Table 3-2. PPG on Aluminum Substrate

Cure Temp. (°C)	Cure Period (min)	MEK (Pass/Fail)	Adhesion (Good/Fair /Poor)	Comp/Stretch (Pass/Fail)* 19 8 lb/in		Hardness (1-4)

Infrared Dry and Cure:						
45-120	5	P	P	F/F	F/P	3
45-120	8	P	P	F/F	F/F	3
45-90	8	F	G	F/F	F/P	2
45-85	10	F	G	F/F	F/F	2
45-75	10	F	G	F/P	P/P	2
45-70	5	F	G	F/P	P/P	1
45-70	10	P	G	F/P	P/F	2
45-70	8					
then at 70	10	P	G	F/P	F/P	2
45-65	8					
then at 65	6	P	G	F/P	P/P	2
45-65	8					
then at 65	7	P	G	F/P	P/P	1
Infrared Dry and Oven Cure:						
150 +-5	10	F	F	F/F	P/P	1
150 +-5	10	F	F	F/P	P/P	1
150 +-5	10	F	F	F/F	P/P	1
185 +-5	15	P	G	F/P	P/P	1
200 +-5	15	P	-G	F/P	P/P	2
220 +-5	15	P	G	F/F	P/P	2

* A pair such as "F/P" gives ratings (here, Fail/Pass) for the inside and outside of a fold, respectively, here and in succeeding tables.

Table 3-3 indicates considerable success with the "Bonderite EP-2" rolled steel substrate, but the Compression/Stretch test must be discounted somewhat because the very thick samples supplied by the manufacturer would not bend into tight creases under the loads we were using. This was compensated to some extent by the fact that the greater thickness led the coating on the outside of the bend to be stretched considerably, and the coating on the inside to be compressed as much. Since the subject of interest is the efficacy of the cleaning and surface preparation process, the results of the Adhesion and MEK tests indicate that it quite successful.

Table 3-3. PPG on Rolled Steel (Bonderite) Substrate

Cure Temp. (°C)	Cure Period (min)	MEK (Pass/Fail)	Adhesion (Good/Fair /Poor)	Comp/Stretch (Pass/Fail) 19 lb/in	Hardness (1-4)
Infrared Dry and Cure:					
45-85	8	P	G	P/P	2
45-85	8	P	G	P/P	2
45-65	8				
then at 65	7	P	G	P/P	3
Infrared Dry and Oven Cure:					
200+-5	15	P	G	P/P	3
220+-5	15	P	G	P/P	3

Table 3-4. PPG on Untinned Carbon Steel Substrate

Cure Temp. (°C)	Cure Period (min)	MEK (Pass/Fail)	Adhesion (Good/Fair /Poor)	Comp/Stretch (Pass/Fail) 19 lb/in	Hardness (1-4)
Infrared Dry and Cure:					
45-65 +-2	8				
then at 65 +-2	7	P	G	P/P	1-2
40-60 +-2	8	P	G	P/F	2
Infrared Dry and Oven Cure:					
200 +-5	15	P	G	P/P	2
200 +-5	15	P	G	P/P	1-2
220 +-5	15	P	G	P/P	2
220 +-5	15	P	G	P/F	2
245 +-5	15	P	G	P/F	2-3

(note: infrared dry temperature for this material was 40 +-2°C)

Sheet steel, both tinned and untinned, gave satisfactory results for most tests, regardless of cure, as shown in Tables 3-4 and 3-5. This material seemed particularly strong in the compression and stress tests. The hardness of the coating remained modest for all cures and substrates, while still showing good resistance to compression and stretching. Its adhesion at lower oven cure temperatures and for infrared curing is quite adequate, and its overall performance at 220 C is quite good.

Table 3-5. PPG on Tinned Carbon Steel Substrate

Cure Temp. (°C)	Cure Period (min)	MEK (Pass/Fail)	Adhesion (Good/Fair /Poor)	Comp/Stretch (Pass/Fail) 19 8 lb/in		Hardness (1-4)
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Infrared Dry and Cure:

40-60+-2	9					
then at 60+-2	6	P	F	P/F	P/P	2-3

Infrared Dry and Oven Cure:

200+-5	15	P	F	P/P	P/P	2-3
220+-5	15	P	G	P/F	P/P	3

(note: infrared dry temperature for this material was 40 +- 5°C)

Copper presents itself as one of the more consistent of the materials; in Table 3-6, little difference can be discerned between the two types of curing. Like the sheet steel, copper attains only modest hardness over the ranges tested.

Table 3-6. PPG on Copper 161 Substrate

Cure Temp. (°C)	Cure Period (min)	MEK (Pass/Fail)	Adhesion (Good/Fair /Poor)	Comp/Stretch (Pass/Fail) 19 8 lb/in		Hardness (1-4)
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Infrared Dry and Cure:

40-55	10					
then at 55	5	P	G	P/P	P/P	2
45-65	8					
then at 65	7	P	G	P/P	P/P	2-3

Infrared Dry and Oven Cure:

225+-5	15	P	G	P/F	P/P	2
220+-5	15	P	G	P/P	P/P	2

(note: infrared dry temperature for this material was 40 +- 5°C)

In conclusion, for the PPG Powercron 600 cationic epoxy, we found it possible to place one mil thick coatings on five different substrates with good resistance to MEK, tolerance for stretching and compression and good adhesion. The coatings were not as hard as we might have preferred, but if they had been they might not have resisted the bending tests as well. No defects, holes or other flaws were found once the correct drying and curing times had been established, and all samples passed the conductivity test until after undergoing the compression stretch test (folding), for which the results are noted in the tables.

3.6 Testing of Glidden 105 Anodic Epoxy

Initial trials with the Glidden 105 black anodic epoxy uncovered several problems. Among these was a tendency for the coating to bubble and become unevenly distributed. Manufacturer's specifications were followed as closely as possible (though this was not always exactly possible), but the problems continued. After considerable experimentation it became evident that it is necessary to treat this epoxy differently from the PPG Powercron 600 epoxy, especially with respect to procedures immediately following removal of the sample from the coating chamber. It was found that excessive rinsing with distilled water immediately after removal from the coating bath hindered clean and uniform drying and that immediate evaporation aided by the infrared lamps produced a more uniform coating.

By its anodic nature, this epoxy is essentially limited to applications on aluminum. Results of tests, specified by dry and cure, are given below. The maximum voltage was set to 120 $\pm$ 2 volts while the current was limited to 45 $\pm$ 10 mA. Coating thicknesses of a little over one mil showed no significant difference between infrared and oven cure.

Table 3-7. Glidden Anodic on Aluminum Substrate

Cure Temp. (°C)	Cure Period (min)	MEK (Pass/Fail)	Adhesion (Good/Fair /Poor)	Comp/Stretch (Pass/Fail) 19 8 lb/in	Hardness (1-4)
Infrared Dry (45°C for 2 minutes) and Oven Cure:					
180 \5	20	P	F	F/P F/P	2-3
180 \5	20	P	F	F/F F/F	3
Infrared Dry (55°C for 4 minutes) and Oven Cure:					
180 \5	20	P	F	F/P F/P	2-3
180 \5	20	P	F	F/P F/P	2
Infrared Dry (45°C for two minutes) and Cure:					
45-60	8				
then at 60	7	P	P	F/F F/F	1
55-110	25	P	P	F/F F/F	3

The above information reflects the strengths and weaknesses of the numerous samples that were made and tested. In general, the hardness that can be attained with this coating is admirable, while adhesion and resistance to compression and stretching is poor to average. In particular, regardless of cure and dry cycle used, this coating was unable to pass either the heavy or the moderate compression test. Since the coatings in an actual absorption chiller might be subject to similar distortions, or to lesser distortion repeated many times as the system flexed, this coating seems less promising than the PPG anodic coating of the previous section, and it was primarily for this reason that less time was expended in investigating its properties.

A second reason for moving on to the Glidden cathodic coating was the expectation that cathodic coating in general is preferable to anodic coating. When a sample serves as the anode, the metal substrate is oxidized to metal ions that become part of the finished coating. The presence of such ions in the coating has detrimental effects on corrosion resistance. In a cathodic system there is no decomposition of the metal being coated, and so there are no residual ions in the coating to lessen corrosion resistivity.

Finally, it should be noted in closing that the possibility of changing the relative concentrations of resin and pigment feed as a way of improving coating characteristics adhesion can not be ruled out, although we did not have time to pursue this avenue here. While the manufacturer's specifications were closely followed for these trials, a higher concentration of resin feed could have a beneficial effect on adhesion. The general brittleness of the coat, however, is a characteristic that would remain unaffected.

3.7 Testing of Glidden 500 Cathodic Epoxy

Aluminum was the material selected for initial testing of Glidden Cathodic Low Gloss Black 500 epoxy. While this epoxy seemed much more amenable to drying and curing than its anodic counterpart, it had an unusual granular texture which was evident immediately after removal from the coating bath. Testing of the surface revealed, however, that the coating contained no pinholes and that it had excellent resistance to methyl ethyl ketone. The value of this coating to our applications does come into question in compression, stretching, and adhesion tests, which gave thoroughly unsatisfactory results, as the tables below indicate. Optimum voltage was much lower than for other coatings, at 120 volts, while current was 15 ma/in².

Table 3-8. Glidden Cathodic on Aluminum Substrate

Cure Temp. (°C)	Cure Period (min)	MEK (Pass/Fail)	Adhesion (Good/Fair /Poor)	Comp/Stretch (Pass/Fail) 19 8 lb/in	Hardness (1-4)
Infrared Dry (45°C for 2 minutes) and Oven Cure:					
170+-5	20	P	P	F/F F/F	2
190+-5	20	P	P	F/F F/F	3
Infrared Dry (45°C for two minutes) and Cure:					
45-70	8				
then at 70	7	P	P	F/F F/F	2-3
45-75	7				
then at 75	8	P	P	F/F F/F	3

This cathodic epoxy seems to share the strengths and weaknesses of its anodic counterpart. The complete inability of the Glidden coatings to withstand compression and stretching makes their applicability to coating thin foils in absorption chillers or other applications minimal at best.

3.8 The Problem of Sharp Edges

Despite our success at laying down smooth, uniform, flexible coatings on copper, aluminum and tinned steel in the 3 to 5 mil thickness range with the PPG cathodic epoxy, there was one significant problem: the coating process left bare sharp corners, such as where the foil had been cut by shears. Sometimes the entire flat edge of the foil (a 1 mil wide surface) would be exposed, and sometimes only the corner itself. In either case this constitutes a significant problem, since corrosion could start here and gradually work under the entire coating. Consultation with manufacturers indicated that this is a common problem, and that the coating is pulled back from the corner by surface tension when it is semi-liquid during the heat cure which follows the actual coating process. Experimentation with different heat-cure cycles produced little improvement.

We did find that the quality of sample edges can be substantially improved by treatment with highly (80-90%) concentrated amounts of the respective cleaners on the areas in question. While it is still difficult to obtain flawless edges, such concentrated applications to the edges reduce exposed metal from 50-80%, at a considerable cost in preparation time.

In sum, we are not satisfied that the edges can be satisfactorily coated by the electroplating process, and think that it may be necessary to add a bead of solvent-deposited polymer sealant on the edges of the heat exchanger after fabrication. Since this would involve only the edges of the metal it would not affect the working heat exchanger area and would involve only minimal amounts of sealant. Identification of an optimal sealant has not been carried out as yet.

3.9 Coating of Interior Surfaces

Many heat exchangers involve both an outside and what could be called an inside - the interior of tubes in a tube sheet, for example. All polymer coating experiments described above involved coating the exterior of a sample, that is, a surface with a near "line of sight" exposure to the opposite electrode through the polymer. Since corrosive liquids may well flow through these interior regions as well, some effort was devoted to attempts to get the coatings to plate out inside tubes.

Since the deposition of the coating involves the flow of ionic current through the solution, against resistance due to the scattering of the ions by water molecules, it was always clear that we could not expect a heat exchanger with long internal passages to be uniformly coated simply by flooding it. The ability of different materials to coat through a given displacement in the solution is called the "throwing power" in industry, and it is measured in inches and feet. We had hoped that a combination of careful arrangement of components and rapid pumping of the coating solution through the sample would overcome this resistance. A series of tests on copper tubes have shown that this is not the case.

A system for depositing polymer coatings on the interior of complex structures such as our heat exchangers was designed. The sample (a piece of tubing) is made an integral part of a hydraulic circuit. The system then pumps the polymer-bearing solution through the sample at high speed while the

voltage pulse is applied; our hope was that any slight polarization of the polymer would be translated by bulk motion into additional deposition current. The coating is, of course, recycled back to the reservoir after it leaves the substrate area.

The pump employed was a Masterflex 7021-24, designed so that the solution only touched the workpiece or the interior of the Tygon tubing used to transport it. The system delivered 2280 ml/minute, which gave a velocity of 66 inches/sec through the 3/16 inch i.d. copper tubes used as test pieces. After coating, the samples were cut open and examined visually and using the conductivity test for coating integrity.

Pumping the solution through the tube produces coatings of 4 to 6 inches length inside the tubes, about what was expected from the manufacturer's specifications for throwing power. However, the coatings were of lower quality than those obtained on exterior surfaces, with some pinholes appearing in 60% of the samples. Virtually all samples demonstrated average to good adhesion, hardness, and resistance to MEK. Changing the position of the workpiece inlet in the bath had little effect. Increasing coating time from ten minutes to one half hour increased the length of the coated area by some 25%, but seemed to endow the initially coated areas with a scaly, uneven texture. Increased voltage up to 300 V produced longer coatings also, but at an unacceptably low rate (about .023 inches/volt). Insertion of a thin wire anode into the tubing, suitably protected from short circuits, was difficult and resulted in very uneven coatings, adequate in some spots, visually incomplete in others. On the basis of these results, the coating of interior regions must be ruled infeasible at this time.

The impact of this failure on system design and feasibility is not great. Essentially it means that we cannot coat the interior surfaces of the evaporator, absorber or condenser (which are exposed to system water); there is no problem coating the external surfaces which are exposed to lithium bromide solution. So, for example, the use of steel or aluminum in the evaporator may now be ruled out since it would be difficult to protect from system water, but the use of copper foils in these applications still looks very promising, and the second effect generator has a simple enough geometry to be coated on both sides if we make it from a metal.

4. EROSION AND CORROSION TESTING OF ELECTROPLATED POLYMER COATINGS

4.1 Introduction

In Section 3 we described our attempts to electro-coat with polymers the interior of tubes and similar structures formed from foils. Had this effort been successful, a major area of interest would have been the rate at which streams of water would erode the polymer coatings, especially at the sharp bends in the stream which would occur at the manifolding points in the foil structures, where the stream flow would lead the water to rush directly against coated surfaces. The impracticality of coating the interior surfaces has rendered this series of tests irrelevant, and they have been omitted.

The exterior surfaces of foil heat exchangers can be coated; here erosion is not a significant concern, since our design calls for relatively slow fluid flow over these surfaces. In some cases, however, the fluid will be lithium bromide brine, and corrosion of the surfaces is a matter of significant concern; in principle the coatings described in the previous section should protect the foils completely, but it is necessary to test this in practice. Due to enthusiastic endorsements of these coatings by knowledgeable experts and on the manufacturer's recommendation, we initially assumed that they would work well at low temperatures (40 or $100^{\circ}\text{F} = 5$ or 38°C) or in pure water and resolved to test their performance in the more demanding environment of, for example, the second effect generator at 100°C ($= 212^{\circ}\text{F}$). We also wanted to know both whether a perfect coating would stand up to the hot brine and whether an imperfect coating - one with an exposed edge or pinholes - would be lifted off the foil by the advance of corrosion under the coating. We therefore immersed various samples in a 100°C brine, as described in the following sections.

4.2 Corrosion Testing of Polymer Coated Samples

Samples of aluminum, copper, bare steel and tinned steel were coated with PPG's cationic epoxy using the best techniques developed during the work described in Chapter 3. The samples were one inch wide and about five inches long, of which 4.0 inches was coated. For each type of foil, samples were prepared in pairs, one member of which was immersed as coated, while the other was painted around the edges and on all uncoated surfaces with a solvent-based commercial anti-etching coating ("Stop-off") which was expected to be impervious to the brine, although it is much too thick to be a permissible coating in a heat exchanger. This painted sample, then, tested the resistance of a coating protected on the edges from brine intrusions, while the "as coated" samples tested the ability of the coating to withstand being undercut by the brine at unprotected edges. The two possibilities are shown in Figure 4-1.

All samples were suspended so that the bottom 1.5 inches was submerged in a stirred 105°C ($= 220^{\circ}\text{F}$) lithium bromide bath. (The top layer of the brine in this open container is the most highly oxygenated and thus the most corrosive.) The upper part of the sample was inevitably exposed to some splashing and because of being in the air was exposed to an even more corrosive environment. The samples were left in the bath for sixteen days, then removed and examined carefully.

Figure 4-1. (a) A coated metal sample. (b) A coated sample painted with "Stop-Off".

At this time, as noted below in discussions of individual metals, some of the unprotected metal samples had been attacked to the point of falling from their suspension. However, none of the samples exhibited any visible pitting or corrosion on the PPG coated regions. There were some harbingers of instability in the Stop-Off coatings in the presence of a dark, viscid substance around the stop-off/polymer border and a clear tendency of the Stop-Off regions to develop pinholes in the more thickly coated upper portions. These samples were then either returned to the tank or replaced by a fresh sample. At this time the samples which lacked Stop-Off had the holes by which they were suspended coated with Stop-Off to lessen the corrosive effects of the string tension.

The samples were then left in the bath for several months, being checked regularly and examined carefully after 35, 49 and 189 total days of immersion. The results are discussed for each sample below, but the clear conclusion is that the coatings cannot be considered reliable at these temperatures ($105^{\circ}\text{C} = 220^{\circ}\text{F}$), in an oxygenated brine since even for the best case (rolled carbon steel) the coating is brittle and cracks when bent after 189 days of hot immersion. We conclude that electrocoated epoxies are not an acceptable option at this temperature range, and suggest that the next step is a series of tests to ensure that the claims of good performance in the 100°F (40°C) temperature range apply to foils immersed in lithium bromide.

The question arises of whether the coatings would have performed adequately in the de-aerated conditions that obtain inside a chiller; that is, was the oxygen responsible for these failures? Certainly the oxygen accelerated the corrosion of any metal to which it had access. We do not know

how large a role oxygen (or lithium bromide, for that matter) played in the observed embrittlement of the epoxy coating, which might be a chemical reaction or might simply be an unwanted continuation of the internal chemical processes that solidified the epoxy in the first place. In either event, this result is troublesome. Since the reason for including the coating is to protect the metals from residual oxygen in the system, the failure to do so at these temperatures is a serious problem, and the oxygen serves simply as an accelerator that allowed us to diagnose the problem more rapidly. Although much more detailed experiments would be needed to establish reaction rates and make a quantitative assertion, it currently appears that if the chiller interior is sufficiently oxygen-free to permit the use of epoxy coating at these temperatures, the metals probably do not need the protection of the epoxy coatings.

4.3 Corrosion of Epoxy Coated Aluminum

At the time of the first examination, after 16 days of submersion, the unpainted aluminum samples had suffered such a great degree of corrosion at its suspension point that the sample became dislodged and lost in the bottom of the tank. The painted aluminum sample, which was still suspended firmly, had been torn apart, perhaps by the agitating rotor in the tank. These samples were replaced, with another painted aluminum being added. This time, however, the unpainted samples had the holes by which they are suspended coated with the anti-corrosion agent to lessen the effects of the string tension.

At the 35 day inspection, the badly corroded unpainted sample was discarded, and it was clear that only samples where the coating edges were protected by paint were worth considering. Even this was becoming doubtful, as bubbles of about one centimeter diameter had appeared in the coating on both samples. By the time of the 49 day inspection the bubbles had gotten quite large and were clearly filled with a vapor (hydrogen or steam) since they were causing the samples to float. Between then and removal at 189 days the rest of the aluminum corroded away leaving the polymer coating in a brittle and useless stage containing a white powder, presumably aluminum salts. Some of this damage may have started at faults in the paint covering the edges, but some of the bubbles clearly started up right in the middle of the electrocoated epoxy, indicating both the existence of an undetected flaw and a marked intolerance to these flaws on the part of the aluminum. It is clear that at these temperatures, epoxy coatings on aluminum foils are not useful.

4.4 Corrosion of Epoxy Coated Copper

At the time of the first examination, after 16 days, the unpainted copper sample, also, had corroded loose from its support and was lost in the tank; it was replaced with another unpainted sample, which also disappeared, so the use of unpainted copper samples was abandoned.

The painted copper samples did not fare much better. Above the average brine level a blue-green deposit formed on the outside of the Stop-Off, indicating copper was migrating through it. At the same time, bubbles containing brine formed on the epoxy coating, sometimes near the center where the edge effects should be minimal. At the final inspection (after 189 days) much of the coating was gone, what remained was brittle and friable, and the

copper foil had been eaten right through in several places. The rest of the copper surface appeared not corroded and had not changed measurably in thickness. For copper foils as well as aluminum, the conclusion seems inescapable that epoxy coatings are not a useful option at these temperatures.

4.5 Corrosion of Epoxy Coated Bare Carbon Steel

At the first (16 day) examination, microscopic evaluation of the unpainted sample revealed what appeared to be a slight raising of the polymer coating away from the edges, little or no deterioration of the exposed metal was observed. There was no discernible deterioration of the painted sample, although a small bubble in the corner of the treated sheet steel sample had developed, apparently due to insufficient Stop-Off on that spot. Since the metal still had not been exposed, the sample was re-touched and put back. Shortly thereafter the unpainted sample disappeared, due to corrosion at its point of suspension.

At the 35 and 49 day inspections there was no discernible deterioration of the bare steel sample, and no noticeable deposits on the outside. When it was finally removed from the brine after 189 days, however, detailed examination revealed substantial oxidation within the apparently intact epoxy coating: the sample thickness had increased from about 5 mils to 30, and there was a layer of dense, red material with crystalline inclusions between the layers of epoxy. Both ferric bromide and ferric oxide have the right general appearance, and we did not attempt to determine which this was. It is also possible that the epoxy was serving as an electrolyte and a new compound was forming directly within it, so that it ended up much thicker than its original one mil. A thin layer of apparently intact metal also remained at some points, immersed in the red material.

Although it would be interesting and perhaps valuable to know why the epoxy coatings did not become embrittled and detached in the case of the bare carbon steel, and how this led to the creation of such a dense adhesive layer, it is also clear that once again the epoxy failed to protect the substrate from the hot, oxygenated brine.

4.6 Corrosion of Epoxy Coated Tinned Steel

The unpainted tinned steel sample was badly corroded (in the uncoated areas) by the time of the 16 day examination, and was replaced by another sample with painted edges and support hole. On both of these samples, the epoxy coating and painted edges retained an excellent appearance at the 35 and 49 day examinations. One of them, however, apparently had been badly painted near the support hole, as it corroded off of its support and had to be retrieved from the brine.

After 189 days, however, both samples had deteriorated markedly. In contrast to the untinned steel, here the epoxy coating had become detached from the steel over large areas and had also become cracked and embrittled, much as the coatings on the aluminum and copper had. Underneath, the steel had rusted, as it would in water, rather than forming the dense red layer. There were spots where the steel had completely dissolved, other areas where brown, flaky oxides remained and a few spots where relatively clean metal

could be seen. Apparently, the tinning lessened the bond between the steel and the epoxy, and the free circulation of brine near the steel produced a quite different corrosion from that which occurred under the high integrity coating on the untinned steel. In any event, it still appears that the epoxy coatings are quite unsuitable for high temperature use in lithium bromide brines.

5. ELECTROPLATED HEAT EXCHANGER FABRICATION

5.1 Introduction and Summary

Our objective in this task was to fabricate a small prototype heat exchanger from polymer electroplated metal foil and test it for heat transfer characteristics. The device was to be made from aluminum, copper or carbon steel foil, electroplated on one or both sides and tested in the system described in Task 6. (Since we have not succeeded in welding steel foils, that material was dropped from consideration.) Construction was to follow the plans and procedures similar to those used in making a polymer boiler, described in Chapter 9. Then, using the same procedures, the heat transfer characteristics of the device would have been measured and, by comparison with the results for the polymer boiler, the relative impact of the polymer coating would have been determined. However, given our finding (see Chapter 4) that the polymer coatings are completely unsuitable for use at boiling temperatures, the motivation for this experiment evaporated and we decided to use our remaining resources in more fruitful areas.

Should we discover, at a later date, that the polymer coatings are suitable (with respect to corrosion and erosion) for use in lower temperature heat exchangers, the next reasonable step would be to coat the outer surface of a set of foil tubes to create a recuperator and test it in comparison with an identical recuperator made of uncoated copper in the test stand described in Chapter 6. Until we have an indication that the coatings can withstand the LiBr environment, however, limitations of available time and funding preclude this next logical step.

6. HEAT EXCHANGER TEST FACILITY

6.1 Introduction

Our objective in this task was to establish a capability for measuring the heat transfer properties of small recuperators to allow us to determine the heat transfer properties of polymer films and polymer-coated foils in these applications. We did this by constructing an appropriately instrumented test bed which maintains a steady flow of hot and cool water through the two legs of recuperator-like heat exchangers and allows temperature and flow data to be acquired. This system, and its operation with simple copper heat exchangers is described in this chapter. The performance of other heat exchangers in this test beds is described in the chapter in which the heat exchanger is discussed.

It is worth noting that in general our instrumentation philosophy was in the end minimalist - that is, we had started with notions of automated, computerized data taking, but in the course of setting up the system and dealing with balky flow meters and noisy connections we realized that setting up a sophisticated system was occupying more time than it would save. Accordingly we switched to using a digital thermometer with a multi-position switch to read the various thermocouples, static manometer columns for measuring pressure, and a stopwatch and large calibrated beaker for measuring flow rates. The consistency of most of our results vindicates our use of this approach.

Figure 6-1. Recuperator test bed. T, p and F refer to measurement points for temperature, static pressure and flow rate.

6.2 Recuperator Test Bed

The recuperator test rig was designed to test polymer or metal foil recuperators at various flow rates and temperature differentials while subjecting them to low static pressures so that materials could be tested for heat exchange properties without great concern for mechanical strength, which was being measured in Task 1. Two tanks, elevated at about nine feet, held hot and cold water as shown in Figure 6-1. The levels in these tanks were held constant by float valves connected to the supply, so the static head was quite constant. Flow was modulated by the ball valves indicated in the figure, and of course this adjustment altered the total pressure in the recuperator being tested. Temperature (from a digital thermometer coupled to type "T" thermocouples) and total (manometer) pressure were measured at the input and output of each side of the recuperator, as indicated by "T" and "p" in the figure, and the flow rate was measured at the exit points from each leg using a stopwatch and calibrated beaker.

The system was assembled and tested using an all copper tube-in-tube recuperator operating in counterflow. After each flow rate was set by adjusting the valves, the flow rate was measured, the pressures observed and then the four temperatures read off. Finally, the flow rate would be measured again. The two flow rates usually agreed to within a few per cent, and all other measurements were equally stable once the system had equilibrated for a given setting of the valves, a process which took only a minute or so. The resulting data for four runs of the copper tube recuperator are shown in Table 6-1, as well as calculated heat balances indicating that the system is operating well and has reached a steady state. The heat balance in per cent, HB, is defined as:

$$HB = 100 * (1 - (F_c/F_h)*(T_{co} - T_{ci})/(T_{hi} - T_{ho})). \quad 6-1$$

Table 6-1. Performance of Copper Tube-in-Tube Recuperator

Run #	F _h (gpm)	F _c (gpm)	T _{hi} (°F)	T _{ho} (°F)	T _{ci} (°F)	T _{co} (°F)	Heat Balance (%)	Effective- ness (%)
1	.986	.627	154	110	77	149	3.9	.916
2	.648	.577	155	98	77	142	1.5	.827
3	.453	.559	154	91	77	133	9.0	.858
4	.383	.559	153	86	76	123	2.3	.880

* Subscripts: h = hot side, c = cold side, i = input end, o = output end.

The effectiveness, E, is the ratio of the amount of heat actually transferred to the largest amount that could have been transferred, had the hot stream been cooled to the incoming cool temperature and the cool stream been heated to the incoming hot temperature. Therefore with F_{min} representing the smaller of F_h or F_c , the effectiveness can be defined either by

$$E_h = (F_h/F_{min})(T_{hi} - T_{ho})/(T_{hi} - T_{ci}) \quad 6-2$$

or by

$$E_c = (F_c/F_{min})(T_{co} - T_{ci})/(T_{hi} - T_{ci}). \quad 6-3$$

These two values would differ by a percentage comparable to the heat balance; the effectiveness, E , given in Table 6-1 is the average of the two.

These data were used to generate Wilson plots (6-1) which graphically demonstrate heat transfer performance. These are based on a simple model giving the total heat transfer, U , as

$$1/U = 1/h_1 + 1/h_2 + 1/h_w, \quad 6-4$$

and asserting that h_1 and h_2 , the heat transfer coefficients associated with the fluid boundary layers, are proportional to the fluid velocity raised to the 0.8 power:

$$h_1 = b_1 v_1^{0.8}; \quad h_2 = b_2 v_2^{0.8} \quad 6-5$$

Thus if one plots measured values of $1/U$ as a function of either fluid velocity raised to the -0.8 power, the result should be a straight line, the intercept of which will give the thermal conductivity of the system if that fluid velocity became so large that the resulting film resistance were negligible. To the extent that this model proves accurate, one can assert that a heat exchanger is behaving normally. Furthermore, the heat transfer coefficient of the wall, h_w , may sometimes be extracted from a Wilson plot of the data and compared to a value based on the bulk thermal conductivity or used to determine the presence of other impediments to heat transfer, such as fouling.

To use this model, one must know both the fluid velocities and the overall heat transfer coefficient U . The fluid velocities are easily obtained from the flow rates given the cross sectional area of fluid flow. For counterflow heat exchangers, with two fluids of equal specific heat C , a well-known relationship (6-2) is

$$E = (1 - Z)/(1 - Z \cdot F_{\min}/F_{\max}), \quad 6-6$$

with

$$Z = \exp(-A \cdot U \cdot (1 - F_{\min}/F_{\max}) / (C \cdot F_{\min})), \quad 6-7$$

where $F_{\min}$ and $F_{\max}$ are the smaller and larger of F_h and F_c . Having measured E , these equations can easily be solved for U .

Figure 6-2 shows a Wilson plot of the data from Table 6-1 for the effectiveness of the copper tube-in-tube recuperator versus flow velocity in the hot leg, and a straight line fitted to the data. The recuperator has a heat transfer area of 1.42 ft^2 and the inner tube (hot side) had a cross section of 0.173 in^2 , while the outer (cold) tube's annular flow area was 0.613 in^2 . The fact that the points lie on a straight line indicates that the model, especially the assumption that film thickness varies with velocity to the 0.8 power, is fairly accurate. The y-intercept of the fitted line, corresponding to $77 \text{ Btu/hr-}^\circ\text{F-ft}^2$, gives U when the hot stream velocity is very large and the corresponding film resistance nearly zero, which is thus the combined resistance of the wall and of the film on the cold side at the observed fluid velocity, an average of 0.304 ft/S . If the cold side has the same slope as a function of fluid velocity as does the hot side, corresponding to $h_h = 13.2 \cdot v_h^{0.8}$, the resulting thermal conductivity of the wall must be

0.065 Btu/hr-°F-ft², far less than the conductivity of copper and indicating fouling of the surfaces. This is not surprising, since one side of the test heat exchanger was in fact quite dirty. The test bed is therefore operational, and capable of providing useful performance data.

Figure 6-2. Wilson plot for copper tube-in-tube counterflow recuperator.

7. ULTRASONIC WELDING OF STAINLESS STEELS

7.1 Introduction

The key to the fabrication of components from thin metallic materials is the ability to weld them. If heavier gauge materials are used - say 50 mils and up, as in conventional absorption machinery - then arc welding, brazing and so on are simple, reliable and well known (although sometimes costly in terms of labor). When welding thinner strips and foils, however, it becomes increasingly difficult to produce reliable bonds or to avoid burning out the materials.

During the BNL solar collector project (7-1,7-2) two solutions to this problem had been identified: laser welding and ultrasonic welding. Laser welding appeared to be intrinsically expensive, a significant defect when cost reduction is the goal. Ultrasonic welding appeared to be cost effective, and had the added advantage of producing minimal metallurgical disruption of the metal, which is cold worked rather than melted and re-crystallized. Thus anti-corrosive properties such as inclusions and grain structure were more likely to be left intact than in the case of laser welding; a brief description of ultrasonic welding is offered in Section 7.2.

We had therefore purchased, adapted and installed an ultrasonic welding machine (Sonobond, Inc., of West Chester PA, Model 2050 welder and power supply). In the collector project we had used this machine extensively on copper foils, and in Phase I of this project (7-3) we had extended its use to aluminum and nickel alloys; this work is discussed in Section 7.3 below. The ability to weld steels, and in particular stainless steels, is an important goal, however, and one which had eluded us until recently; in Section 7.4 we discuss our previous (failed) attempts to weld steel foils.

As a result of a re-analysis of the welding process we then tried an aluminum oxide coating on the the weld wheel and discovered that we were able to make consistent, leak-free and fairly strong welds in 316 stainless steel foil. We regard this as a major success, with the potential for many applications in areas other than absorption chillers or even heat exchangers; it is described in Section 7.5. We also attempted to extend this technique to tinned and bare 1010 carbon steel, but found the steel to be too brittle to form reliable bonds. We were unable to obtain suitable samples of various "superalloy" stainless steels to see if they could also be welded. These applications of the aluminum oxide wheel are discussed in Section 7.6.

7.2 The Process of Ultrasonic Seam Welding

The essence of ultrasonic seam welding is indicated in Fig. 7-1. A metal disk, the "weld wheel", rolls without slipping across the two pieces of foil which are to be joined. Its path will be the seam. The head is not hot; welding takes place because the head is vibrating along its axis at an ultrasonic frequency, and this vibration is transmitted down into the pieces of foil. The resultant churning of the metal results in a breakup and subduction of the oxide layers at the adjacent surfaces of the two pieces of foil, and when the completely clean metal surfaces are exposed to each other they are free to weld spontaneously. (This welding is normally inhibited by the oxide layers, dirt and other impediments which are here cleared away by

the vibrations.) The result is a bond of approximately the width of the weld wheel edge in which it is normally impossible to observe any trace of the old surfaces; the cold working is obvious when looking at the seam from either side, and the side on which the wheel was pressing will normally have a track pressed into it for a depth of some fraction of a mil.

There are three dynamic parameters which must be adjusted to give an optimal weld: the power being transmitted through the wheel as a high frequency (about 20 kHz) mechanical wave, the "clamping force" with which the weld wheel presses against the work piece and the speed with which the wheel rolls along the workpiece. For a given material and foil thickness, these must be adjusted to find the particular values that give maximum weld strength. The wheel speed is usually limited by control requirements. Although the requisite power must increase with wheel speed, the relationship is not simple linearity, as might be guessed; also, for a given speed and pressure, going beyond the optimal power produces weaker welds, either through thinning of the foils or excessive cold working of the material. Finding an optimal set of parameters can be a bit tedious, but once found they are very repeatable for a given material and foil thickness.

Figure 7-1. Ultrasonic Seam Welding - Key Components.

7.3 Ultrasonic Welding of Non-Ferrous Materials

Certain metals are easily welded together ultrasonically, using a tool steel weld wheel. These include copper, aluminum and nickel alloys. Table 7-1 shows the strength of the welds as a function of various parameters for Copper 151 (a corrosion-resistant zirconium alloy). All points are averages based on tensile tests with a hand held force gauge of three narrow strips from different points along the same seam. Note the decline in strength at high powers at lower speeds and the extreme power requirements implied for high speed operation. Table 7-2 indicates the relative lack of sensitivity to total clamping force.

Table 7-1
Strength of Ultrasonic Seam Welds in Cu 151 (lb/inch)*

Power(W)	Head Speed (in/min):				115	137	159
	27	49	72	93			
50	27	40	33	35	27	18	8
75	53	42	42	46	0	8	0
100	53	70	37	31	0	4	0
150	72	63	47	38	38	28	18
200	57	61	56	46	44	35	36

*Weld wheel clamping force of 46 lb for all tests.

Table 7-2
Strength of Seam Welds in Cu 151 (lb/inch) versus Clamping Force*

Force on Head(lb):	26	34	42	50	57
Weld Strength (lb/inch)	54	59	57	62	66

*Power = 150 W, wheel speed = 44 inches/sec.

Similar tests on 3 mil foils of 3003 Aluminum and M-400 Monel resulted in weld strengths of 25 and 42 lb/inch, respectively (7-3). No further work was done with these or other metals except as noted below.

Concern may arise as to the integrity of the welded seam. A definitive test would employ evacuation of a chamber connected to a helium leak detector, with the foil arranged so that air pressure would be tending to tear the weld open rather than close it. Lacking time and resources for this level of testing, we sealed acetone in an envelope by welding four seams around the edges of two squares. Despite repeated heating (which produced expansion and distortion of the packet) and other rough manipulations the packet lost no measurable amount of acetone (less than 1 mg out of about 1000mg) over a period of six months.

We have also demonstrated that it is quite feasible to weld three layers of copper foil together at higher powers (300 W), with weld strengths

of 55 lb/inch. These welds were found to be equally likely to break on either the weld wheel side or the table side. This may prove to be very useful in heat exchanger design. We also successfully welded 3 mil foils onto massive blocks of copper, a key step in the construction of larger, more complex structures.

7.4 Previous Attempts to Weld Steels Ultrasonically

Welding the materials discussed in Section 7.3 proceeds with remarkably few problems. This is not always the case, and there are (at least!) three ways in which ultrasonic welding can fail: the material of the foil being welded can stick to the weld wheel, the two foils can fail to bond at all, or the welding process can in some way weaken or damage the foils so as to render them useless. All of these problems have turned up one way or another in our attempts to weld steel foils. For various purposes in this project, we were interested in the possibility of welding foils of 316 stainless steel, of the various corrosion-resistant "superalloys" discussed in Chapter 8, and of ordinary 1010 carbon steel, tinned or bare. It is noteworthy that bronzes, which are copper alloys containing tin, are difficult or impossible to weld ultrasonically.

Our first attempts focused on 316 stainless, following on earlier efforts reported elsewhere (7-1,7-2) and outlined below. In these earlier efforts the welds have either been non-existent or seriously flawed and steel has been deposited on the wheel, effectively destroying it, within the first few feet. This was first discovered using an ordinary carbon steel weld wheel on 316 stainless. The wheel welded itself to the stainless, tearing the foil to pieces.

Since it was clear that the problem was that welding was taking place between the wheel and the workpiece, the problem appeared to be one of finding a material from which to make the wheel, or with which to surface it, which would not be compatible with the steel workpiece. Several coatings were tried. The first was Haynes alloy 90. It was arc welded onto the surface of the wheel and dressed to size. It was found to produce welds only intermittently, and the tip surface deteriorated rapidly.

At that point we were searching for hard surfaces which could be firmly attached to the weld wheel. We turned to the leading vendor of surface hardening coatings, Union Carbide. The first we tried, on their advice, was LW-1N30, which is detonation gun bonded Tungsten Carbide. This is 87% tungsten carbide and 13% cobalt, blasted onto the steel substrate (the rim of the weld wheel) at very high speed from a gun-type device. Its primary attribute is listed as extreme resistance to wear and a very high "tensile strength" - that is, resistance to removal from the substrate, once applied. A total of four weld tips were coated and tested over a few months, with various surface preparations. In most cases no welds were observed, although 5% of the seam was welded at certain control conditions (250 Watts, 30 lb clamping force and 62 feet per minute). Adding oil between the weld tip and the top foil, and then between the foils raised this to a maximum of 50% of the seam being welded. However, the weld wheel was becoming coated with steel and deteriorated rapidly to the point where it could not be used. In other tests of LW-1N30 the coating chipped off the weld tip, leading to rapid deterioration.

Union Carbide then recommended their WT-1 coating. This is also a detonation gun bonded surface, consisting of Tungsten-Titanium Carbide (83%) and Nickel (13%). This coating is as smooth as the surface it is deposited on. Tested with lubricating oil (as above) about 10% of the seam was welded, but coating deterioration was apparent before the seam was completed. Raising the power to 300 Watts led to the rapid deposition of the coating onto the foil.

7.5 Aluminum Oxide Weld Head

At this point we reviewed the problem carefully. The carbide coatings were bonding to the steel foil and this was pulling them off the wheel. They had very high tensile strength ratings, but this did not help. If we had a coating which was less likely to bond to the steel foil, wouldn't that be more likely to stay on the wheel? Union Carbide offered such a coating, called LA-2, a Detonation Gun-applied ceramic, Al_2O_3 . They had not recommended it previously because its tensile bond strength is lower than the carbide coatings, but crystalline Al_2O_3 is sapphire, one of the most inert substances known, and we hoped that its strongly covalent/ionic (i.e. non-metallic) chemical bonds would minimize adhesion to the metal foil.

Union Carbide then coated a suitably prepared weld wheel with the LA-2 aluminum oxide coating. Microscopic examination showed the surface to be extremely rough, like sandpaper, so we expected there would be no slippage of the weld wheel on the upper foil, which may have happened with the WT-1 coating. (Slipping is undesirable, since it produces energy dissipation at the wheel-foil interface, with less energy being transmitted to the foil-foil interface below, where it is needed.) Low power tests indicated no sticking of the 316 stainless steel to the weld wheel, but no welding either. As the power was increased to 100 and 150 W, however, we saw striking success. Very quickly, we found that we had successfully welded several feet of seam welds in 316 stainless steel with no noticeable deterioration of the wheel. Further tests demonstrated that the wheel has not deteriorated visibly under microscopic (60X) examination after hundreds of feet of welding and we have no reason to doubt that this coating can be used in a production mode.

In the course of these tests we determined the optimal parameters (wheel pressure, power and speed) for welding our 3 mil 316 stainless foil. As for copper, dependence on the clamping force was very weak, and we normally use 50 lb, as for copper. About 60 inches per minute is a convenient speed, and the dependence on power under these conditions is shown in Table 7-3 for two different experiments. These results make it clear that too much power can have a very deleterious effect on weld strength. It is also clear that these welds have only one third to one-half the strength of copper, at best. This is unfortunate, but we have not found a way to adjust the parameters to improve the situation. Examination under an electron microscope showed that there is no visible thinning of the material, and that there is no residual boundary between the foils in the welded zone, so the cold working must be making the steel more brittle than it was when annealed. These strengths are adequate for many of our needs, so this relative weakness should not be a cause for undue alarm.

Table 7-3						
Strength of Seam Welds in 316 Stainless Steel (lb/inch)*						
Power to Load (W)		75	100	150	200	250
Sample	Tests/seam					Speed (in/min)
#1	2	22	28	18	17	14
#2	3	25	23	11		

*Clamping force = 46 lb.

We have also discovered (Section 7-6) that the temper of the metal is extremely important to welding success; cold rolled samples of 304 and 316 stainless steel which have not been tempered are almost impossible to weld; it may be that samples which are annealed to be even softer than our standard roll of 316 will be capable of stronger welds than those reported here, but this will have to be tested at a later date.

The appearance of the welds after they have been torn apart is quite striking. Under a microscope they can be seen to consist of a dense series of parallel beads which often, but not always, overlap. At first it was not clear whether these beads were parallel to the rolling striations in the foil or to the head motion, but a series of experiments where welds were made with the seam first parallel and then perpendicular to the striations, and where the foil pieces were aligned with striations perpendicular to each other, clearly demonstrated that the beads follow the axial vibrations of the weld wheel.

The presence of these beads, and in particular of the unwelded regions between them, raise the same questions about the integrity of these seams as were expressed for copper. Accordingly, we sealed a small quantity of acetone in a four-sided envelope seam welded from 316 stainless steel, heated it, cooled it and tested for mass loss. As for the copper, no significant loss was detected, but under time pressure this envelope was only tested for about two weeks. As for copper, a careful test with a leak detector should be set up when possible.

7.6 Welding Specialty Stainless Steels and Carbon Steel

Our success with 316 stainless led us to try to extend our ability to weld steels to various specialty stainless steels known for their high resistance to corrosion and discussed in our Phase I report and in Chapter 8 below. The materials include AL6X, AL6X+N, 29-4C and others. We were unable to obtain samples of commercial foils made from these materials, since commercial demand is too low to sustain a market and minimum job size is too large for us to be able to afford to pay for specialty rolling.

We consequently set about preparing foil samples at BNL from 1/8" thick stock obtained from the Metal Samples Corporation. First these were rolled out to 3 to 4 mil thickness. Attempts to weld these samples were complete failures - the foil still did not stick to the weld head, but no bond was formed between the two foil pieces at all, at any power or pressure.

The samples of foil were obviously much harder than the commercial 316 stainless steel we had been working with, as could be recognized by look, feel and sound. (In fact, the first failure was with a sample of 316 stainless rolled at BNL for comparative purposes, so we know the problem did not lie in the presence of the extra components that give the specialty alloys their corrosion resistance.)

We then tried to temper our newly rolled foils but found that our equipment was not capable of carrying out the job. To prevent oxidation of the foils, a hydrogen atmosphere is a requirement during the annealing process. However, successful annealing of these materials also requires a rapid quench when the heating cycle is over. Our ovens were unable to satisfy these requirements simultaneously and produced either untempered steels or charred, brittle black residues. Since the specialty stainless steels are in most ways metallurgically similar to the 316 stainless we have had success with, we have no reason to doubt that the technique will work, but it will not be possible to demonstrate this under the current contract.

We also attempted to extend our ultrasonic welding to 1010 carbon steel foil, to increase our flexibility and design options. We tried both 2.5 mil bare carbon steel and, despite the known problems with tin-bearing compounds, we tried a 2 mil tinned steel foil, which has much higher corrosion resistance than the bare foil. In both cases the welding failed, although in a new way. Again there was no problem of adherence of the steel to the weld head - it still looks as it did when new. Between the foil pieces very weak welds would form, at a wide variety of parameter settings, which would separate under very light forces. After separation, the weld seam on the foil away from the weld wheel would be seen to be perforated by tiny holes, and (under the microscope) the foil next to the weld wheel would have (on the side facing the other foil) tiny bits of foil which had been ripped out of the holes. The shapes of holes and bits were irregular and matched perfectly. This happened almost identically on both the tinned and untinned foils, and we were unable to prevent it by any parameter adjustment save turning down the power until no welding occurred at all. Although it almost certainly involves some aspects of work hardening under the action of the weld head, exactly why this peculiar form of breakdown occurs is a mystery to us, and is the subject of ongoing research.

8. LITHIUM BROMIDE CHILLER CORROSION MECHANISMS

8.1 Introduction

Our goal in this task was to determine the corrosion mechanisms most likely to occur in alloys used in a thin materials chiller and to identify materials which would make possible the construction of a thin materials chiller. To make a chiller of thin metal foils, at most a few mils thick, almost all corrosion must be eliminated if reasonable service life is to be obtained. We believe we have established that this is possible for several superalloys for use in the first effect generator (especially AL6-X) and that crevice corrosion will not be a problem for copper foils in the absorber, condenser and evaporator, although there is still a possibility of pitting corrosion in these components which must be examined at a later date.

There are many aspects to the design of low cost absorption chillers that differ significantly from the more conventional systems where information, although little quantitative data, on the corrosion behavior of many metals has been obtained. It was therefore necessary to identify differences in the design and to conduct short term tests to determine if any major increases in corrosion would take place with the thin wall components. In view of the limited time available for this study, rapid testing was carried out where virtually no information was available from the literature.

Copper and steel have been used successfully in chillers and data on the behavior of carbon steel, types 304 and 316 stainless steels, nickel, cupronickels and Monel is available (8-1,8-2). In general, most metals, including copper, corrode uniformly in LiBr, except for 70-30 cupronickel which showed shallow pitting when air and 10% NH_3 were present at 200°C. When stressed, stainless steel showed stress corrosion cracking if air was present, but this cracking was inhibited when LiOH was added to concentrated bromide solutions.

The above results are important with regard to the behavior of metals in the bromide environment. The use of superalloys should reduce the corrosion rates below that of stainless steel if they are not susceptible to various forms of localized corrosion, since stress corrosion cracking can be prevented by increasing the pH of the solutions. Measurements were therefore carried out to determine the susceptibility of a series of superalloys to localized corrosion. The presence of localized corrosion either in the form of pitting or crevice corrosion would be extremely detrimental to the use of foils and could be a rapid form of their failure.

Crevice corrosion was also considered a possible mode of corrosion of ultrasonically welded copper in the coolant waters. The narrow gaps present along the welds form tight crevices and if crevice corrosion could take place this would limit the life of the copper heat exchangers. There was no crevice corrosion information available for copper, and as the metal is susceptible to pitting in certain environments accelerated measurements were carried out on copper crevices to determine their corrosion characteristics.

During tensile tests of polymers in LiBr failure of stainless steel support rods occurred where they had been contaminated by LiBr. The failure analysis carried out on these rods, which showed characteristics of stress corrosion cracking, will also be presented.

8.2 The Corrosion Susceptibility of Superalloys

In the work of the previous phase (8-1), several metals were identified that could be used in the various components. The first effect generator is exposed to a gas flame on one side and to a boiling film of lithium bromide solution on the other, at about 300 °F. It must almost certainly be made of a specialty stainless steel, such as AL6X, AL6X+N or 29-4C, on the basis of tests in the previous phase, and here we refine this conclusion and establish the suitability of these materials. The second effect generator is exposed to condensing water on one side and boiling Lithium Bromide solution on the other and operates at about 190 °F. We believe this can be made from a polymer or from a mild steel foil, but if not, the same list of candidates might be of interest.

The localized corrosion behavior of metal and alloys which owe their corrosion resistance to the presence of a passivating surface oxide layer can be assessed using electrochemical techniques. When the metal is polarized with an externally applied potential, breakdown of the passive surfaces occur over a narrow potential range above which rapid corrosion takes place. In practice this potential may be reached if the freely corroding potential of the metal reaches this value. Dissolved oxygen both increases the potential and accelerates corrosion after breakdown of passivity and for these reasons should be prevented from entering the solution. (This is the direct reason we have elected not to use polymers, which are inherently permeable, in any heat exchangers containing system water, which would have dissolved oxygen in it; see Chapter 2.)

The eight alloys studied are shown in Table 1 with their compositions. In general the corrosion resistance of the alloys increases with increases in chromium and molybdenum, and nickel reduces the susceptibility to stress corrosion cracking. For test purposes, the alloys were mounted in an epoxy resin, abraded down with 600 grade silicon carbide paper and the edges were masked with a stop off compound to prevent crevice corrosion.

Table 8-1 Composition of Alloys Tested

Alloy	% Composition										Cost*
	Cr	Ni	Fe	Mo	Cu	C	Ti	Nb	Mn	Al	\$/ft ²
Incoloy 825	21.5	42	30	3	2.3	.03	.9		.5	.9	4.80#
Inconel 625	21.5	61	2.5	9	-	.05	.2	3.6	.25	.2	6.40
20Cb-3	20	33	Bal	2.5	2.5	.06	-	8xC	2	-	4.20#
Al6X	20	24	Bal	6	-	-	-	-	-	-	2.20
29-4	29	-	Bal	4	-	.01	-	-	-	-	1.55
26-1	26	-	Bal	1	-	.03	-	-	-	-	2.70#
17-4PH	16.5	7	Bal	-	4	.09	-	-	-	1	2.60#
410	12.5	-	Bal	-	-	.15	-	-	-	-	1.70#

* Cost of 10-mil foil; #Component-based estimates

Testing was carried out in two bromide solutions containing 55 and 60 weight percent of lithium bromide, at temperatures of 135 and 153°C just below their boiling points. The apparatus, a standard electrochemical cell and potentiometer, were described in detail in Ref. 8-1 and are shown in Figure 8-1. Measurements are taken of the current through the sample as a function of the potential of the sample relative to that of a calomel reference electrode at room temperature, starting at -500 mV. The rate of potential increase was from 1 to 10 mV/s. After breakdown had occurred, indicated by a rapid increase in current, the scan direction was reversed. The scanning was usually repeated to determine the degree to which the breakdown in corrosion had repassivated and the subsequent rates of corrosion. The critical breakdown potential was taken as the value when the first sustained increases in current were detected. This was necessary as some alloys showed increases in current followed by a sudden decrease indicative of repassivation of the breakdown sites even though the potential was still increasing. The results of these tests are shown in Table 8-2.

Figure 8-1. Electrochemical cell and potentiostat (schematic).

The measurements of the breakdown potential of the alloys showed many aspects of corrosion behavior. For example, Incoloy 625 showed the highest breakdown potentials for sustained high currents. (See Table 8-2). However, it also showed many transients because of repassivating pitting especially at lower scan rates. The transients may be indicative of a greater tendency for pitting corrosion than the alloys which gave lower breakdown potentials. A similar tendency was shown by alloy 29-4 at 150°C.

Table 8-2
First and Successive Breakdown Potentials (mV) in LiBr Solutions*

Concentration(%):	55	60	60
Temperature(°C):	143	150	150
Scan Rate (mV/s):	5	1	5
Alloy:			
Incoloy 825	10, -50	120, 180	120
Inconel 625	-110	0, -100	-20, -150
20Cb-3	-40, -300	-140, -40	-380, -200
Al6X	-50, -300	-100, -240	-80, -180
29-4	-180, -150, -115	-230, -160	
26-1	-220, -200	-290	-270, -200, -180
17-4	-350, -350	-240, -260	
410	-400, -400	-410, -500	-420, -420

* Measured relative to standard calomel electrode.

Alloy 29-4 also showed an increase in breakdown potential with repeated cycles. This was also observed with Alloy 26-1 but with alloy 17-4 having lower alloy additions, the tendency was for the breakdown potential to decrease with each cycle. The latter characteristic suggested that there was a tendency for general corrosion following breakdown with the lower alloy concentration as was shown in Ref. 8-2 for Type 340 stainless steel. With the higher alloy concentrations, the resistance to breakdown increased after susceptible pitting sites were removed.

Most of the other alloys with breakdown potentials below -0.25 V showed general type of corrosion once breakdown occurred rather than showing localized pitting. Pitting would be a more dangerous type of corrosion than general attack in thin walled structures.

It appears that an alloy with an intermediate chromium/molybdenum concentration such as AL6-X would decrease the possibility of pitting but still reduce the general corrosion. The intermediate alloy concentrations would probably also give faster decreasing rates of general corrosion with time than that was found in Ref. 8-2 for stainless steels. In addition they would reduce material costs relative to the high nickel alloys.

8.3 Crevice Corrosion of Copper in Cooling Water

The second effect condenser/absorber has system cooling water running through it at about 100°F . On the condenser side water condenses from vapor; on the absorber side a film of Lithium Bromide solution absorbs water vapor. These are currently made from copper tubes; we would like to use ultrasonically welded copper foils instead. The evaporator has system cooling water running through it at about 45°F and has a film of pure water boiling off of it. Aluminum is entirely too risky with respect to corrosion, even at these low temperatures, so we would like to use copper foils here also. For both of these components, corrosion due to system water is as much a matter of concern as are the effects of the brine, especially because the system water

is more variable and unpredictable than the hermetically sealed brine.

Copper tubing has been used extensively to carry water containing low levels of inorganic salts. In some waters pitting of copper takes place and has been reviewed by a number of authors (8-3, 8-4, 8-5). In certain waters the pitting takes place within the first year or two after exposure. After this period the pitting rate decreases and failure is less likely. Crevice corrosion has many similarities to pitting, as corrosion products concentrate within a protected volume where convection cannot take place and the concentrated solution is not washed away. In pitting of copper a solid corrosion cap forms over the pit and involves a complex chemistry depending on impurities in the water. With crevice corrosion the formation of this cap was assumed not to be a requirement as the geometry of the crevice itself can prevent convective dilution of the corrosion products. The ultrasonic weld of thin copper sheets forms a very tight crevice between the sheets next to the actual weld and tests were carried out to determine whether the concentration of the corrosion products would accelerate the dissolution of the copper. In the case of iron and nickel and their alloys, rapid corrosion of the crevice region occurs when they are polarized anodically allowing the corrosion products to build up. Little or no corrosion of the freely exposed surfaces takes place if the potential is held within a range where corrosion can proceed but below which general corrosion is prevented. A similar approach was adopted for the study of copper.

The potential of general corrosion of copper was determined from polarization measurements. The most aggressive anions are generally chloride and sulfate and these were used to simulate cooling waters.

Copper wires were polarized at room temperature in solution containing range where copper dissolution took place was above -100 mV. From the shapes of the polarization curves it was apparent that the build-up of copper ions in solution reduced the rates of dissolution. The dissolution rates at the higher potentials decreased with time and deposition of the dissolved copper resulted when the potential was again decreased. Similar tests were performed on copper foils with and without small zirconium alloying additions. After the measurements the edges of the foils showed the most dissolution. The rates of dissolution were greater in the sulfate than in the chloride solutions. The diffusion rates at the corners and edges were the most rapid and here the build-up of the dissolved copper offered the last protection to the foil.

Marked differences in the cathodic reduction of oxygen were also noted in the sulfate and chloride solutions. In the chloride solution the reaction was markedly catalyzed under certain conditions when only a thin oxide was present on the metal. This behavior disappeared when the oxide thickened after the metal had been exposed for a period of days.

The protection by the corrosion products was again important when welded foils with narrow or opened crevices were tested at a fixed potential where dissolution of the copper would be expected. Little corrosion of the crevices took place. The concentration of copper ions in the crevice again led to protection rather than corrosion in the crevices. Dissolution of the edges again took place but was not representative of what would occur in practice, being rather a result of the accelerated nature of the test.

A scanning electron microscopy study of the corrosion sample was carried

out. In Fig. 8-2 the edge of a sample held at a fixed potential for three days showed extensive dissolution of the edge of the sample and precipitates toward the crevice. In general two corrosion products were formed. The first was a uniform layer over most of the metal surface except where rapid dissolution took place; the second was a crystalline precipitate. Both appear in Fig. 8-2.

Figure 8-2. Dissolution of Cu Edge.

Figure 8-3. Welded Crevice Opened.

Fig. 8-3 shows a tight crevice which has been folded open to reveal the crevice and show the location of the precipitates. Some precipitates were also formed away from the weld where the metal surfaces were in close proximity forming a crevice. In the crevice close to the weld precipitates can be seen mirrored on both metal surfaces.

The crystals grew mainly as platelets. This can be seen at the higher magnification in Fig. 8-4, taken close to the weld. The layer of corrosion products over the surface can also be seen in this figure where it had been cracked on bending open the foil. At the outer areas (away from the weld) with regions of precipitates, the uniform deposit was apparently less protective. Disruption of the oxide cover can be seen at these specific sites in Fig. 8-5. This may have been the first signs of pitting or simply a transition between the protected and unprotected regions, but because of the limited period of testing a clear distinction could not be made.

Figure 8-4. Crevice at high magnification

Figure 8-5. Platelets far from crevice.

In conclusion, this study showed that crevice corrosion did not develop in a manner that would be expected with iron and nickel alloys and will not be a mode of failure with the ultrasonically welded copper foils. Further investigations to determine whether the crevices would act to assist pitting corrosion should be undertaken because of the indications of this possibility.

8.4 Failure Analysis of Stainless Steel Rods

During the creep tests of polymers described in Chapter 1, the stainless steel rods supporting the polymers failed under very low loads. The rods were at an intermediate temperature between the solution test temperature and ambient (probably about 90°C). At and above the level of the bromide solution extensive rusting had taken place. After the exposure, the rods were bent where the rusting had occurred and fractured easily, suggesting stress corrosion cracking had taken place below the rust. Below the bromide level there were no signs of corrosion. It was therefore concluded that access to oxygen was the important variable.

The fracture surfaces were studied in the scanning electron microscope and it was clear that stress corrosion had taken place. The investigation also showed that the stainless steel rods had large amounts of sulfide

inclusions indicative of a Type 303 stainless steel. The high concentrations of sulfur possibly increased the susceptibility to stress corrosion cracking. This observation points up the need to select an alloy resistant to stress corrosion in any external structures (exposed to air) where unexpected spills or contamination may occur. An alloy would not crack in this manner in the de-aerated bromide, since oxygen is necessary to the process.

8.5 Corrosion Considerations for other components.

The two recuperators, exposed only to lithium bromide solution, are now made of mild steel plate. We will probably recommend that they be made of a polymer (PEEK; see Chapter 2), but if a way is found to extend ultrasonic welding to steel foils their low cost would make them strong candidates. There will also be structural elements, including a base plate, frame and top plate, which will be reasonable thick to lend structural support, but which will be exposed to bromide solution. Strength requirements indicate they must be made of a metal, but whether it should be mild steel or a stainless is unclear; corrosion information may make the difference in this choice, and the Ref. 8-2 and 8-6 will be useful sources of this information when design decisions are actually made.

9. GENERATOR DEVELOPMENT

9.1 Introduction

Our objective in this task was to examine the feasibility of constructing a miniature generator consistent with our baseline design, and if possible, to build and test it. Several designs and materials are relevant and of interest, and with sufficient resources we would have pursued several of them. Under the constraints of time, however, we were able to test only a PEEK generator, operating at 212°F. Such a device could well serve as a second effect generator in our design, although probably in another orientation, and this demonstration that nucleate boiling occurs in the normal manner, and measurement of the associated thermal conductivity, furthers the demonstration of the feasibility of our thin materials design.

9.2 Background

Heat transfer during boiling occurs at various points both in traditional absorption machinery and in the thin materials designs studied here. Since we have proposed the use of several exotic or unusually thin materials in these devices, it seems advisable to determine the effectiveness of heat transfer during boiling for these materials.

For the metals a case can be made that although they are thin, their surface interactions with water or brine will be the same as for standard tubing, since the metal surface is physically the same whether the metal beneath it is 5 or 50 mils thick, and the thermal resistance of the metal is negligible in both cases. Accordingly, when it became clear that we did not have time to conduct all the tests we would like, we focused on boiling from a polymer surface, since this process is central to the operation of our proposed second effect generator, relatively unknown (see the discussion in Chapter 10 below) and since there is reason to fear that the smooth, low surface energy characteristics of the polymer film might inhibit nucleation. For simplicity we boiled pure water, since the extensive testing reported in Chapter 1 assured us that PEEK would withstand the LiBr environment without problems at these temperatures.

9.3 Design and Fabrication

We constructed the simple apparatus shown in Figure 9-1, consisting of a PEEK bag filled with heat transfer oil, an electric heater and several thermocouples. The bag rested in pure (distilled) water containing another rack of thermocouples just above those in the oil. The PEEK bag was of the simplest possible design, four inches wide and twenty inches long. It was welded shut on the two long sides and one end and bent up at the open end to retain the oil, keep the water out and allow entrance to the wires. It had to be held down by a simple wire frame, since the oil had a lower density than water. The two heaters were nine inches long, leading us to use a heat transfer area of one half a square foot in our calculations. The two heaters rested side by side, two inches apart, and the two lines of thermocouples were actually between and slightly above them, not directly above them.

Figure 9-1. Boiling heat transfer test apparatus.

The water was contained in a clean open box welded from PEEK and resting inside a box made of one inch polyurethane foam board. A rectangle of PEEK floated on top of the water, covering almost all of its surface and inhibiting heat loss through evaporation while permitting us to observe the boiling below. The upper row of thermocouples rested about one quarter inch above the PEEK surface.

The only operational problem encountered was a tendency for the oil to produce an unidentified gas at the surface of the heaters, which would be trapped under upper surface of the PEEK bag and inhibit heat transport. We kept these bubbles away from the thermocouples and manipulated them out when they became troublesome.

9.4 Testing

Since the most power we applied was normally 230 Watts (to minimize the oil breakdown), the system heated slowly. As it heated, convection became apparent both in the oil and in the water. Then the water began to de-aerate, forming tiny bubbles at numerous sites on the bag surface. As the indicated temperature of the water passed 200°F and that of the oil passed 240°F, steam bubbles began to form on the bag and break free, re-condensing immediately. With indicated temperatures of 210°F many bubbles were forming and breaking free, only to re-condense while trapped under the PEEK cover. At 212°F (water temperature) the boiling was too intense to see what was happening below. It was clear, however, that substantial boiling was also taking place under the bag, as large steam bubbles came rushing up the sides of the box.

Figure 9-2. Temperature difference versus heat transfer for PEEK boiler.

The important qualitative observation is that there was no shortage of nucleation sites; bubbles formed freely at numerous points, and as the oil temperature increased, the number of sites increased, rather than having the added heat removed through more vigorous boiling at existing sites. This indicates that the surface characteristics of PEEK are qualitatively satisfactory.

The observed temperature differences at the three different thermocouple sites are shown as a function of applied power under conditions of stable boiling in Fig. 9-2. The ordinate is scaled in units of overall heat transfer, assuming that the power is uniformly distributed over the entire bag. This cannot, of course, be the case; there will be less heat transfer at the ends, for example, and indeed, T_2-T_5 is the largest temperature difference. Nonetheless, the data indicates that for this arrangement we could expect temperature differences on the order of 40°F at the 65 Btu/min-ft^2 we expect to flow through our second effect generator. Current temperature differences in the second effect generator are on the order of 30°F (Appendix) and the LMTD in our computer model (Chapter 11) is 22°F . If the enhancement that condensation on the hot side will produce (compared to the oil) is taken into account, we have every reason to expect that PEEK will operate perfectly satisfactorily as a boiler or generator.

10. CONTROL OF PHASE CHANGE PROCESSES

10.1 Introduction

Our objective in this task was to determine the feasibility of ensuring drop-like condensation and nucleate boiling through surface selection and preparation, and to assess the overall heat transfer enhancement which would result from this. Our plan was to rely primarily on the collation and assessment of existing knowledge and to examine the possibility of developing advanced polymer electroplating techniques. Our studies have led us to conclude that we can expect substantial benefits from the use of PEEK (or other polymers) wherever condensation occurs and the polymer can be used. With respect to specifically enhanced nucleate boiling the situation is less clear, primarily because the problems we have had with polymer coatings have left us with few options for surface treatments. We have established (in Chapter 9) that the use of a polymer surface seems unlikely to cause problems, and that boiling will proceed from a PEEK surface much as it does from a suitable metallic surface.

10.2 Potential of Condensation Applications in Chillers

It has been widely known since early in this century that heat transfer involving condensation can occur in either of two modes, drop-like or film condensation, and that the thermal resistance is much lower for the case of drop like condensation. In coarse summary, if a surface is hydrophobic, leading the condensate to bead up and then run off quickly, there is no thermal resistance from the film of water some fraction of a millimeter thick which forms in the other case. Considerable effort has consequently gone into finding suitable coatings which will make (metallic) condenser surfaces hydrophobic, starting long ago with various oils which inevitably washed off after a few hundred hours. We will quote only the work of two currently working groups.

The first group, at the U. S. Naval Postgraduate School, have been engaged in an extensive study of the effects of polymer films on boiling and condensation (10-1). Their results compare the enhancement of heat transfer due to drop-like condensation, which is made possible by the polymer coatings, to the increased thermal resistance of the coating itself by measuring steam side heat transfer coefficients. Their results indicate that although most of the coatings do enhance drop-like condensation, only those that can be laid down with thickness substantially under one mil result in significantly enhanced heat transfer. A Soviet group (10-2) arrived at the same conclusion in a study of a PFA Teflon film, recommending films only if thicknesses below 0.15 mils could be maintained.

Recalling that our initial goal was to obtain the corrosion-resistant characteristics of the polymer films without paying a price with respect to heat transfer, these results would appear to indicate that this cannot be the done with the coatings we have examined to date, since our electro-deposited polymer films were 1.0 to 1.2 mils thick, and we found that even that thickness was not sufficient for corrosion protection. Also, if an oxygen-free environment is maintained inside the chiller, the condenser tubes experience only pure water and are in little need of corrosion protection, so the coatings must justify their cost solely on the basis of heat transfer. It

may be possible to achieve this with some of the very thin coatings examined in Ref. 10-1, and the possibility should be examined in the future.

Drop-like condensation has been observed at large heat fluxes on an all polymer (PFA Teflon) condenser constructed to operate in an unpressurized mode (10-3). BNL staff have witnessed this device in operation and we regard it as a positive demonstration that polymer film heat exchangers will perform efficiently in condensing heat exchange. Of course there is a residual temperature drop from the material, but the extraordinary strength of PEEK makes it possible to reduce this dramatically through lower thickness film. The remaining step is to demonstrate that PEEK will produce the same level of beading and run-off that PFA Teflon does. This demonstration should be carried out, but because the hydrophobic nature of PEEK is just as apparent as is that of Teflon, we are quite confident of the result. The unpressurized design of this particular device renders it inapplicable in absorption machinery.

10.3 Potential of Boiling Applications in Chillers

The possibility of enhancing boiling on polymer-coated surfaces through the creation of permanent nucleation sites is also of interest. The Soviet group mentioned above have also tested heat transfer into a lithium bromide solution through steel tubes coated with a polymer for corrosion protection (10-4), and found very substantial thermal resistance (comparable to the liquid phase resistance) from a 5 mil coating and more modest resistance from a .6 mil coating. This is as expected, but raises the hope that by enhancing nucleate boiling by altering the surface of the polymer one could compensate for the added thermal resistance.

We had intended to dust a polymer-electroplated heat exchange surface with various proprietary powders which then be frozen into place during the curing process. We anticipated that the rough corners of the dust would harbor permanent bubbles of steam or air which could serve as nucleation sites, enhancing boiling considerably. We still think this idea has promise, but have postponed experimentation until we can find a coating which will better withstand the hot brine.

It is also known that surface treatments which enhance the wettability of the evaporator or absorber can increase performance dramatically, in particular on copper and cupro-nickel surfaces (10-6). These techniques are in common use commercially, and would certainly be applied to copper surfaces in a thin materials chiller as well, especially a "falling film" design, where it is extremely important to encourage spreading of the film.

11. MODELING CHILLER OPERATION

11.1 Introduction

Our objective in this task was to prepare a computer simulation model with which to assess the overall feasibility of our baseline design and to determine the performance impact of design choices. To do this we surveyed existing simulation models of absorption chillers and selected the recently completed "ABS" model developed by Dr. G. Grossman for Oak Ridge National Laboratory as the most suitable and flexible.

We then went through several steps to prepare the model so that it would stand as an accurate representation of our conceptual designs for a thin materials chiller. First we adapted the model to be an accurate representation of the "best current practice" design for a 25 ton chiller developed during Phase 1 of this effort. Having thus calibrated the model, we adjusted the parameters representing the sizes and connections between the various heat exchangers to match the requirements of our conceptual design, based on concentric cylinders. Under a variety of qualifying assumptions, we are able to predict from calculations using this model that this design would operate with a gross COP of 0.99 (including stack losses) and a net COP of 1.32.

This chapter also contains a description of our conceptual design, describing some the functional relationships, assumptions and remaining undefined details.

11.2 Background

Computer simulation is now a widely accepted tool in engineering design, allowing many alternative systems to be compared and evaluated without the expense of actually developing many different prototypes. Systems such as TRNSYS (11-1) for solar energy, DOE-2 (11-2) for building energy consumption and hundreds of other proprietary and non-proprietary special purpose programs have been used to model systems ranging from tiny pumps to electric utilities. We wanted be able to predict the performance of a thin materials chiller with some degree of credibility, and accordingly examined the range of available models. In some cases sophisticated models had been prepared to examine the dynamic effects (11-3) occurring when chillers were starting up; since we had decided to focus first on design point operation these were eliminated. Many other special purpose models had been constructed (11-4,5), including one at BNL (11-6), but all of these lacked the flexibility that would permit us to compare the design alternatives we were interested in on a direct basis.

The ABS model (11-7), authored by Dr. Gershon Grossman, appeared to meet all our requirements, since it was modular in form, allowing one to plug a selection of components together in an almost infinite number of permutations and aimed exclusively at modeling design point steady state performance. The model had the further advantage of being compatible with advanced personal computers. We obtained a copy of both the Fortran code and a compiled and ready to run version along with many example data sets from ORNL and set to work.

Fig. 11-1. Block diagram of a standard series coolant flow double effect chiller.

11.3 Model Validation

To learn the intricacies of the model, compare it with hand calculations and assess the validity of our starting point, we first ran ABS to describe the "best current practice" 25 ton double effect chiller (hereafter, the "BCP chiller") presented in Tables 3-3 and 3-4 of our Final Report on the first phase of this project (11-8). These tables are reproduced here as an appendix. This was a directly fired double effect chiller designed to run at high LiBr concentrations to ensure sub-atmospheric operation at all times. (This simplifies code compliance and makes design much simpler, since forces will always be in the same direction on a given piece of material.) A block diagram of this design is shown in Fig. 11-1; details are in the appendix and Ref. 11-8, where the component sizes and operating conditions (temperatures, pressures, heat flux, etc.) are all specified and derived.

The ABS model is very flexible with respect to how many and which variables are used to specify a chiller configuration, as long as the system is not so over-specified as to be unrealizable. We defined the system in a manner analogous to constructing a machine, and then let the model tell us how it would run. We did this by specifying the heat exchanger sizes (the heat transfer coefficient times the area, or "UA's"), and the values used are specified in Table 11-1. They were chosen so that the log mean temperature differences (LMTD's) calculated by ABS would agree with the temperatures in Tables 3-3 and 3-4 of Ref. 11-8, with a certain amount of trial and error fitting of the results.

The chiller capacity was specified as 25 tons by the 45°F temperature of system water leaving the evaporator, the 55°F temperature of system water returning to the evaporator, and the 60 gpm flow rate. The cooling water was specified with an 85° return temperature from the cooling tower, corresponding to the wet bulb temperature on the design day. The ABS model had no specific capacity for modeling the fire box of a directly fired chiller; we evaded this problem by specifying that very hot water (450°F) was flowing through the first effect generator, producing appropriate changes in solution temperature over the heat exchange surfaces of a direct fired system, as specified in Table 11-1. For analysis, we continued to assume 25% flue and jacket losses in calculating the difference between net and gross COP.

Pumping rates for the weak solution and cooling water are specified for the BCP chiller, as well as flow rates for the other streams. We could not specify any of these rates as fixed using ABS without over-determining the system and making it impossible to find solutions. (ABS solves a set of equations, representing conservation of mass and energy and other applicable physical relationships, for a set of unknown variables, such as temperatures and concentrations. The number of variables must be equal to the number of equations; if there are too many variables the system is underdetermined, if there are too few, it is overdetermined. In either case, no solution can be found.) We therefore left them as variable, let ABS find values as part of the solution process, and will assume that appropriate pumps can be purchased to match these flow rates and make the system work as specified. All of the flow rates determined by ABS were within 20% of those we would have specified.

The brine concentration was determined by requiring the weak solution to have a concentration of 56.0% LiBr by weight and letting ABS determine the

other concentrations, with the results shown in Table 11-1. It would clearly have been preferable to specify 57.0% LiBr by weight for consistency with the BCP chiller, but ABS was unable to find feasible solutions at higher concentrations, where we tried 56.5 and 57%. Since we know that the machine will work at 57% (see discussion in Ref. 11-8), this results from some inaccuracy in ABS which we did not have time to track down. The condition of sub-atmospheric operation was maintained, but the observed small drop in COP may stem from this difference.

The result was quite good agreement between the ABS model and the BCP chiller, as shown in Table 11-1 where heat transfer for both cases is shown for each heat exchanger. The heat transfers are not defined exactly the same in the two models, but the heat exchangers do seem to be working significantly harder in ABS, and the condenser a bit less hard, as explained below. The net COP from Ref. 11-8 was 1.33, while that obtained from ABS was 1.32, most probably due to the decrease in brine concentration, while most temperatures agreed within a degree or so. This agreement is close enough to conclude that our BCP chiller and the ABS version agree quite well with each other and this lends credibility to both calculations.

Table 11-1. ABS Simulation of "Best Current Practice" 25 Ton Chiller

	--Best Current Practice ¹ --		----- ABS -----	
COP (Net and gross):	1.33, 1.00		1.32, 0.99 ²	
Concentrations(%):	57.0, 60.0, 62.0		56.0, 58.5, 60.7	
Component	Area (Ft ²)	Heat Transfer (Btu/min)	UA (Btu/min-°F)	Heat Transfer ³ (Btu/min)
1st Effect Generator	50	3750	27	3940
2nd Effect Generator	65	2830	135	2955
Condenser	75	2750	480	2458
Evaporator	100	5000	625	4950
Absorber	150	6000	420	6430
Hi Temp Recuperator	100	3080	190	3620
Lo Temp Recuperator	100	1930	110	2120

1) BCP chiller From Ref. 11-8, Tables 3-3 and 3-4.

2) $COP = (\text{heat removed from output stream}) / (\text{heat removed from input stream})$

3) Heat transfer in ABS = $UA * (\text{Log mean temperature difference})$.

The residual differences may have arisen from a few engineering details that could not be easily programmed into ABS in addition to the difference in concentrations; for example, according to Ref. 11-8, the intermediate solution undergoes a temperature drop of 7°F in leaving the first effect generator due to flash in the separator (i.e., rapid evaporation after the solution has left the region of the heat source) and there was no way to include this effect, which is dependent on the detailed design of the generator, into ABS. The error was simply distributed among the state points adjoining the generator, leaving some two or three degrees above the values in Ref. 11-8, some two or three degrees below. This was sufficient to allow the recuperators to operate more effectively than in Ref. 11-8, leaving less work for the second effect generator and condenser.

11.4 Baseline Thin Materials Design

We here present a preliminary conceptual design for a chiller manufactured from thin materials. Much of the preliminary thought comes from Ref. 11-8, and is not repeated here. Similarities to the Batelle "falling film" design (11-9) will also be clear. The basic design is shown in Figure 11-2, and consists of a collection of concentric cylindrical structures. Pressures are such that all the thin films and foils comprising the heat exchange surfaces are under tension. Thus the first effect generator is a simple cylinder made of AL6-X stainless (see Chapter 8), with weak solution dripping down the outer surface of the inner cylinder and boiling out into the innermost annular space. The heat source is presumed to be from extremely uniform gas combustion within the inner cylinder, probably sustained by a Pyrocore fiber matrix combustor (12-10) or equivalent, producing predominantly radiative heat transfer, enhanced by reflective end caps at the top and bottom of the annular space between the combustor and the first effect generator. Much of the development of falling film boiling, and indeed, how much is boiling and how much rapid evaporation (which is clearly preferable), remains to be worked out, as do possible surface treatments or stamped-in flow-modifying surface irregularities.

The resulting vapor condenses on the inner side of the second cylinder, which is the second effect generator. We are specifying PEEK for this component because of its immunity from corrosion, demonstrated ability to withstand the conditions and its hydrophobic character, which will encourage drop-wise condensation. (See Chapter 10.) Thus a splash screen between the first effect and second effect generators may be unnecessary, since an occasional drop of brine on the PEEK will not promote corrosion.

Intermediate solution is then distributed on the interior of this PEEK cylinder and is boiled and evaporated off by the heat released by the condensation on the other side. For the area and heat transfer of our BCP chiller, the temperature drop across two mil PEEK would be 3.7°F, far less than the 44°F temperature drop in the BCP design, or the 22°F drop calculated by ABS for that design. Two problems with this design have the same solution: because the cylinder is large (see Table 11-2) PEEK is not strong enough to withstand the stress due to the one psi (or so) pressure increment from the first to the second effect generator. Also, because it is hydrophobic, the brine will tend to bead up and run off quickly. Both problems can be attacked by adding either windings of stainless steel or polymer wire, or a mesh of an appropriate material, to take up the stress and continually redistribute the falling brine. We have indicated this in the figure by including a set of concentric wire hoops, but at this point the exact form of the reinforcer/distributor is an open question.

The next cylinder out is a sheet of parallel vertical tubes formed from two sheets of copper foil by ultrasonic welding. (Other geometries might prove advantageous and we present this arrangement only as an example.) The inner surface serves as a condenser for vapor released by the second effect generator, while the outer surface is the absorber, with dense brine dripping down it. Here, also, surface treatment and irregularities may be useful in slowing the flow of the absorbent film. System water flows through the tubes, so the absorber and condenser are cooled in parallel, rather than in series as in the best current practice design.

Figure 11-2. Thin materials chiller - conceptual design.

The final working surface is a copper foil layer which serves as the evaporator, bowed inwards on a rigid frame by the external atmospheric pressure. Again, surface treatments and embossing may be important to control the flow of the film of refrigerant. A second layer of foil, ultrasonically welded onto it, carries the system water. Outside this is a layer of insulation and a final, protective shell. If the insulation is built up with layers of reinforcement, such as Fiberglas cloth, the protective shell need not be overly heavy either.

The recuperators will be similar, coiled spirals of counterflowing tubes at the bottoms of the annular spaces in the first and second effect generators, blow molded from welded PEEK films for corrosion resistance and flexibility. The material from which to construct the frame and base plate is still an open question, with steel and reinforced structural polymers both possible.

We developed a preliminary set of dimensions by assuming an overall height of eight feet for the chiller. Assuming, for this preliminary effort, that the thermal conductivities, or "U" values, would remain the same as in the BCP chiller, we determined diameters for the set of concentric cylinders which would give reasonable agreement between heat exchanger areas for the thin materials design and those of the BCP design, given in Table 11-1. The result is the set of diameters shown in table 11-2, along with heat exchanger areas and expected UA values. As shown in Fig. 11-2, we assumed that the two generators and the condenser are only six feet high, with the space below them occupied by the recuperators. For the absorber and condenser, the area is given not just by the height times the circumference of the cylinder, but incorporates an additional factor of $\pi/2 = 1.57$ due to the presence of many small tubes. For the bottom two feet the cooling water is only cooling the evaporator. Note that the only major differences between the two designs are the oversized condenser and evaporator in the thin materials design.

Table 11-2.
Design parameters for thin materials chiller conceptual design.

	-- BCP/ABS --		-- Thin Material Design --			
	Area	UA	Height	Diam.	Area/Vol	UA ¹
1st Effect Generator	50	27	6.0	2.70	50	27
2nd Effect Generator	65	135	6.0	3.50	65	137
Condenser	75	480	6.0	3.80	115	720
Evaporator	100	625	8.0	5.00	125	785
Absorber	150	420	8.0	3.80	150	420
Hi Temp Recuperator	100	190	-	-	1.7 ²	190
Lo Temp Recuperator	100	110	-	-	3.9 ²	110

- 1) Area in ft², UA in Btu/min-°F, height and diameter in ft, volume in ft³.
2) Volume which recuperator may occupy. See text.

Since the recuperators are not cylinders, their areas cannot be calculated in the same way. Instead, we assumed that they have the same one hundred square feet as in the BCP chiller, and ask if there is any problem

stuffing them into the annular spaces at the bases of the first and second effect generators. The volumes available are calculated and shown in Table 11-2, and since they correspond to surface to volume ratios of 60 ft²/ft³ or less, which can be satisfied by tubes of 0.4 inch diameter or less in a shell or a variety of other geometries, we see no fundamental problem here. We have left detailed design questions for a later phase of the work.

11.5 Modeling Baseline Thin Materials Design

The next and final step was the adaptation of the ABS model described in Section 11-3 above to the baseline conceptual design of Section 11-4. We adapted ABS to model the parallel flow indicated in Fig. 11-2 and in the new schematic, Fig. 11-3, by specifying input cooling water temperatures of 85°F for both components and requiring that the cooling water temperatures leaving the two components be equal. We changed the UA values to those derived in Table 11-2 for the thin materials design and ABS then predicted the operating characteristics shown in Table 11-3.

It is clear that there are no significant differences in operation between the two chillers; in particular the parallel cooling and oversized components have had little effect. This result does depend on the assumption that the U values appropriate to the BCP chiller could be carried over to the unchanged. As we gain more certain knowledge of these U values, it will be easy to re-run ABS and determine the operational implications of the new values.

Table 11-3. ABS Simulation of Thin Materials Conceptual Design

	----- ABS/BCP -----	----- Thin Materials Design-----		
Net (Gross) COP:	1.32 (0.99)	1.32 (0.99)		
Brine Concentrations(%):	56.0, 58.3, 60.4	56.0, 58.4, 60.4		
<hr/>				
Component	Area (Ft ²)	Heat Transfer (Btu/min)	Area (Ft ²)	Heat Transfer (Btu/min)
<hr/>				
1st Effect Generator	50	3940	50	3930
2nd Effect Generator	65	2955	64	2910
Condenser	75	2458	75	2480
Evaporator	100	4950	127	4950
Absorber	150	6430	158	6400
Hi Temp Recuperator	100	3620	100	3690
Lo Temp Recuperator	100	2120	100	2020

Figure 11-3. Thin materials chiller (parallel coolant flow) - block diagram.

12. ECONOMIC ASSESSMENT

12.1 Introduction

Taking the conceptual design presented in Chapter 11 as a baseline, we here estimate the expected costs of producing a 25 ton chiller using thin materials technology. Much of the cost data is drawn from the final report on Phase I of this work (12-1), especially Chapters 2, 3 and 5 and, as there, we find a considerable amount of uncertainty inherent in the estimates, in part because the costs of today's smaller machines seem to be much higher than can be readily explained on the basis of materials and labor (12-2).

Nonetheless, a 25 ton thin materials chiller can probably be built for a total materials cost of \$3380, an estimated labor cost of \$3450, and sell, after markups, for a total equipment cost of about \$11,000, or \$440/ton. This is close to the breakeven cost goal established in our Phase I report of \$360/ton or a total of \$9000 for a 25 ton machine (12-3). It misses that goal because of costs not associated with heat exchangers, which may also be susceptible to cost reduction, but which were not specifically studied here. Thus we find that there is a real chance of meeting this cost goal and establishing a marketable chiller. The analysis leading to this conclusion is summarized in Table 12-1 and detailed below.

Table 12.1 Equipment Costs for a 25 Ton Thin Materials Chiller

Component	Area (Ft ²)	Material ¹	Thick- ness (mils)	Unit Cost (\$/ft ²)	Material Cost (\$)	Fabrication ² Cost (\$)	Total Cost (\$)
1st Effect Generator:	50	AL6-X	10	2.20	110	150	260
2nd Effect Generator:	65	PEEK	2	1.00 ³	65	100	165
Condenser & Absorber:	300	Copper 161	3	0.25	75	250	325
Evaporator:	320	Copper 161	3	0.25	80	250	330
Recuperators High Temp:	100	PEEK	2	1.00	100	150	250
Low Temp:	100	PEEK	2	1.00	100	150	250
Base and Head Plates&Frame:		Mild Steel	-		200	300	500
Pumps (Two):			-	-	750	-	750
Controls&Wiring:					800	300	1100
Purge, piping, insulation, shell:					500	700	1200
LiBr, leak test, final assembly:					600	1100	1700
Totals:					3380	3450	6830
Estimated Cost to consumer, includes distributor's markup: 11,000-14,300							

- 1) AL6-X is a proprietary stainless steel; PEEK is Polyether-ether-ketone; SS is stainless steel.
- 2) Includes 400% labor burden; fabrication costs are very uncertain.
- 3) Estimate of bulk purchase price.

Installation costs would not differ significantly from today's machines, since the plumbing, distribution and control systems would not differ significantly from those now in use. The lightness of the thin materials design might lower transportation costs, but not greatly; greater standardization might simplify installation, but this credit would accrue to any new design or marketing initiative. Consequently, we will not consider possible differences in delivery and installation costs further. We do note that it is impossible to make anything low cost with dealer mark-ups of 200%, which now occur in parts of the chiller market. If distribution costs cannot be streamlined through standardization or other measures, there will be little point in making chillers with low factory costs. Operating costs are discussed in the final section, but also would not differ radically from current costs; careful chemical maintenance would still be necessary to control corrosion and fouling.

12.2 Materials Costs

The materials costs are derived from various commercial sources, and are often taken from Ref. 12-1. They are all based on assumptions of large lot purchases at advantageous times, corresponding to the best reasonable prices. Many of them are quite volatile, of course, with copper prices falling in recent years as an example. The cost of PEEK depends drastically on its commercial acceptance; the resin price is only \$0.23/ft² mil, but we had to pay \$250 for 100 ft² of 1.9 mil material. We have taken \$1.00/ft²-mil as a price we are sure can be realized now through bulk purchases.

In some cases it is not certain that the assumed materials will be used. For example, other copper alloys, such as the more expensive cupronickel, might be advantageous in the condenser/absorber if pitting corrosion turns out to be a problem. If PEEK film turns out to be more expensive than we anticipate, copper foils might also be used in the recuperator or second effect generator (until we can weld mild steel ultrasonically it is incompatible with a thin materials design), although the higher temperature parts of the devices would be pushing into a temperature range where the copper foils might creep. In both of these components we think PEEK will be preferable because of the greater flexibility possible in forming complex shapes from it and because of its complete immunity from corrosion.

It is noteworthy that the items on the bottom four lines of Table 12-1 contribute over one half of the material costs to our design. Except for assuming that \$150 worth of "structural" items could be allocated to pay for exterior insulation and a mild steel cylindrical shell, we have simply taken these costs over from Ref. 12-1 without change, since cost reductions in these areas were not within the scope of this project. The arithmetic makes it clear that these items are now the dominant price boosters, and they deserve considerable attention in any future work in this area.

12.3 Labor Costs

The labor costs indicated in Table 12-1 are extremely uncertain but, we believe, conservative. The basis for deriving them was a component by component comparison with the labor costs for our "Best current practice"

(BCP) chiller (Ref. 12-1 and appendix). In each case we compared the complexity of the BCP design to the thin materials design, for example by estimating the number of welds to be made in each case. Thus the great simplicity of the first and second effect generators resulted in substantial cuts in their estimated labor costs. We also took into account the compatibility of ultrasonic welding and seam welding of plastic films with automated production, compared to the large number of brazed joints that are now carried out with a large component of manual control.

As with the materials costs, the fabrication costs are now dominated by items from the bottom of the list which were not part of our purview in this project. It is clear that any steps that can be taken to automate or simplify final assembly and testing will be well worth the effort, and that if these costs are not also brought down absorption chillers will remain a relatively expensive item.

12.4 Markups

In Chapter 3 of Ref. 12-1 we estimated that dealer markups would amount to 66 - 110% on top of manufacturing costs, resulting in a BCP chiller price of \$16,000 to \$20,000 for a 25 ton machine. When we actually surveyed prices, we found \$25,000 to be more representative. We were not able to trace the source of the divergence, since the costs to specific manufacturers were not available to us, but this example makes it clear that if absorption chillers are to become an economically attractive item, something along the distribution chain will have to be streamlined.

12.5 Scaling Up to Larger Machines

Having looked at expected costs for a 25 ton machine, it is tempting to scale up to larger machines, both because there is likely to be demand for them and because one expects "economies of scale" to lower unit prices. We have resisted this temptation for several reasons. First, we think it dangerous to extrapolate from an uncertain beginning point, and our estimates for the 25 ton machine are very emphatically estimates. We think the only reasonable way to estimate costs for a larger machine would be to repeat the entire process for that machine, and we lack the resources under this contract to do that.

Moreover, there are reasons to think that the thin materials design is well adapted to systems made up several small units rather than one big one. Height becomes a problem in many interior installations, for example, since the 25 ton system is eight feet tall already. The larger the system, the greater the absolute forces induced by atmospheric pressure and the more conservative the design must be, especially the space frame. Finally, absorption machinery suffers in efficiency when operated off design point, and a set of machines that can be cut in and out to match a varying load will provide far more efficient operation than will one large one; this principle is already being widely applied in advanced heating systems.

So although we certainly do not rule out the eventual application of thin materials technology to 100 ton or even larger machines, we do suggest

that development focus first on more modest sizes, in the 25 ton range, and that development of large machinery be postponed until this has proven itself.

12.6 Operating Costs

Operating costs are still quite uncertain, as a result of the many undefined element in a thin materials design. We previously (12-4) estimated operating costs of a 25 ton chiller to be about \$1.70/hour for gas and \$0.30 - 0.60/ hour for electricity, depending on electricity prices. Absorption chillers were thus able to compete well against compression equipment in regions of high electricity prices, where a 25 ton compression machine would cost \$3.40/ hour, but fared poorly where electricity was cheap, where compression machines could cost as little as \$1.70/hour.

Two major factors which we have not examined could help the situation. First, if the pyrocore burner allows greater than 75% efficiency in the first effect generator, that could have a large effect, since the price of gas is the dominant variable item for absorption chillers. Second, more efficient pumps, perhaps with DC motors and computerized controls, would lower the cost of electricity, perhaps dramatically, if flow rates which are now controlled by valves and therefore wasting energy were controlled by pump speed instead.

Finally, fixed cost maintenance expenses, which are considerably smaller than energy costs, are unlikely to decrease in a thin materials design. It is clear that the key to these designs is the prevention of corrosion, and it is equally clear (Ref. 12-1, Section 2-4) that the prevention of corrosion is best carried out by careful, frequent chemical monitoring and adjustment of the working fluid and system waters. This is appropriate maintenance for an existing machine, and we see no reason to expect the cost to rise or fall appreciably.

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Appendix 1. A Proposal to Build Heat Exchangers
from Filled, Cross-Linked Polyethylene Films

by. A. Ravve

This is a proposal to consider building heat exchangers from crosslinked polyethylene homopolymer or copolymers. It is further suggested that the plastics be filled with graphite. The proposal is made on the basis of the following considerations. These polymers crosslink when subjected to ionizing radiation. Crosslinking results in formation of chemical bonds between individual polymeric molecules and formation of three dimensional networks. When the temperature of the crosslinked polymers is raised above T_g and external stresses are applied, the chains can no longer be forced to slip past each other and the material is no longer subject to creep. Radiation crosslinked polyethylene is utilized in various commercial applications where dimensional stability at elevated temperature is required. An example of this technology is polyethylene in coatings for cables and wires, where the polymer is crosslinked by irradiation after extrusion onto the cable. The second suggestion to fill these plastics with graphite is based on the thermal conductivity of graphite which approaches that of metals and because the material is not affected by most chemicals, including brine. It appears reasonable to expect that composites of polyethylene with graphite (40-50% graphite) will be better heat conductors than polyethylene alone. Although this does not apply directly, it has been reported that when carbon black is used as a filler in irradiated polyethylene, it reinforces the composite considerably¹. Copolymers of ethylene with fluorocarbons, like Tefzel from du Pont Co. and Halon from Allied Chemical Co. might be good candidates. Both copolymers were reported to crosslink upon irradiation and to be tough and high melting. One must expect some degradation in the halocarbon portions of the molecules as fluoro-carbons are known to undergo chain scission rather than crosslink. How much that affects the properties of the finished products is not clear. Pellets of both materials, filled with graphite, are available commercially. Both materials are much higher priced, however, than polyethylene itself. It appears, based on statements in the literature, that crosslinked high molecular weight polyethylene might actually, be adequate for this task all by itself. Whether this is really so, would have to be determined experimentally.

The units can be manufactured from plastic sheets which can be welded into the desired form. After they are completely assembled, they can then be irradiated. Both types of irradiation may be possible: electron beams from Van der Graaf accelerators or gamma rays from radioactive cobalt or strontium. Electron beams, whether they are scanned beams or electrocurtains are usually easier to use on flat sheets.

The background information on the properties of irradiated polyethylene can be summarized as follows:

It was reported as early as 1952 by Charlesby² that when polyethylene is irradiated with more than 0.1 units of pile radiation (1 unit is equivalent to total flux of 10^{17} thermal neutrons/cm²) the polymer crosslinks. The crosslinking reaction can be illustrated as follows:

The physical effect of chemical crosslinking is small as long as only a few molecules are involved, but increases rapidly as crosslinking density increases. The gel point is reached when a three-dimensional network first extends throughout the system. The dose in rads to reach the gel point³ is expressed by

$$r = (6.023 \times 10^{23} \times 100) / (2 \times 6.24 \times 10^{13} \times G \times M_w)$$

where G is the number of crosslinks produced per 100 eV absorbed and M_w is the weight average molecular weight. A process known as "end-linking" should also be considered. When scission occurs in the main chain of a molecule, at least one of the fragments may link to the main chain of a neighboring molecule to give a branched molecule of a higher molecular weight. The physical properties of an end-linked polymer would be similar to those of a crosslinked one. Figure 1 shows the gel fraction in irradiated polyethylene as a function of the dose in megarads⁴:

Figure 1. Percent Gel Fraction in Irradiated Polyethylene⁴

If chain scission takes place simultaneously with crosslinking, the dose required to reach the gel point is increased. The rate of scission may be measured independently of changes in crosslinking by using the stress relaxation method of Tobolsky⁵ where the sample is stretched to a constant elongation and the retractive force τ is measured at regular intervals. Under equilibrium conditions, τ decreases as the chains undergo scission. The following relation has been derived for g , the number of scission introduced per gram:

$$g = -(1/M_c) \ln(\tau/\tau_0)$$

where τ_0 is the starting tension. If chain scission predominates over crosslinking, the eventual effect of irradiation is weakening the polymer and making it brittle.

After increasing radiation doses, the melting temperature of polyethylene steadily rises until a point is reached where the polymer no longer melts. The crosslinked samples exhibit rubber like elasticity when heated above 120°C. The elastic modulus increases with increasing doses of irradiation⁶. In addition, the polymers exhibit a significant drop in oxygen permeability with irradiation greater than 10⁶ rads⁷. The most outstanding effect of radiation

on polyethylene is the increased heat resistance and form stability at elevated temperatures, considerable reduction in cold flow, stress-cracking and heat cracking⁶.

Irradiation of polyethylene causes an evolution of gas, mainly hydrogen (shown in the above reaction), but also containing a small percentage of hydrocarbons. The hydrocarbon fractions consist mostly of two, three and four carbon compounds which probably come from side chains.

Some of the earliest investigators found that any vinyl and vinylidene unsaturation present in the low density polymers disappears rapidly during irradiation. A more important reaction which accompanies irradiation of polyethylene or any paraffins is the formation of some trans-vinylene unsaturation. This necessitates incorporation of anti-oxidants into the polymer.

The effect of irradiation on creep is well illustrated by a graph in Chapiro's book⁷ showing percent elongation of irradiated polyethylenes which were subjected to a constant load at various elevated temperatures. A portion of this graph is reproduced in Figure 2. It shows percent elongation of crosslinked polyethylenes under constant load as a function of temperature¹⁰:

Figure 2. Elongation versus temperature under constant load.

Curve A	6.7 megarads
Curve B	3.1 megarads
Curve C	unirradiated

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Appendix 2: Typical Characteristics of 25 Ton Absorption Chiller Heat Exchangers

Component	Material ¹	Geometry	Pressure Drop (psi)	Parasitic Power (hp)	Surface Area (ft ²)	Quantity (lbs)	Material Cost (\$)	Fabrication Cost Labor & Burden (\$)	Total Cost (\$)
1st Effect Generator	SS	Shell	0.5	--	50	75	650	300	950
2nd Effect Generator	MS	Tubes or Flatplate	10/0rif	--	65	120	175	350	525
Condenser	COP	Tubes	3 ²	See Abs'r	75	130	290	190	480
Evaporator	COP	Tubes	4 ²	1/2-3/4	100	200	375	275	650
Absorber	COP	Tubes	4 ²	3/4 - 1	150	300	560	450	1010
Hi Temp Recuperator	MS	Flatplate	2.0	--	100	180	75	350	425
Lo Temp Recuperator	MS	Flatplate	2.0	--	100	180	75	350	425
Generator Vessel	MS	Round	--	--	--	200	60	100	160
Absorber Vessel	MS	Round	--	--	--	300	90	150	240
Pumps, Sol'n & Ref	SS/MS	Semi-Hermetic	--	1/4+1/10	--	--	750	--	750
Controls, wiring, etc.						--	800	300	1100
Purge, piping, structural						500	500	700	1200
LiBr charge, leak test, final assembly:							600	1100	1700
							<u>\$5000</u>	<u>\$4615</u>	<u>\$9615</u>
Estimated Cost to Customer; includes Distributor's markup								\$16,000 - 20,000	

¹SS = Stainless Steel, MS = Mild Steel, COP = Copper.

²Water Side.

Appendix 2: Typical Functional Requirements for 25 Ton Absorption Chiller Heat Exchangers

Component	Heat Transfer (mBtu/hr)	Design Pressure (mm)	Entering Conditions				Leaving Conditions			
			Flow Rate ¹	Temp. (°F)	Concen- tration ²	State ³	Flow Rate ¹	Temp. (°F)	Concen- tration ²	State ³
1st Effect Generator	225 ⁴									
Solution		725 ⁵	67.6	277	0.57	l	64.9	305	0.60	l
Refrigerant		-	-	-	-	-	2.7	305	-	v
2nd Effect Generator	170									
Solution		60 ⁵	64.9	177	0.60	l	62.7	194	0.62	l
Refrigerant (HP)		725	2.7	300	-	v	2.7	240 ⁶	-	l
Refrigerant (LP)		0	-	-	-	-	2.2	186 ⁷	-	v
Condenser	165									
Refrigerant		60 ⁵	4.9	186 ⁷	-	v	4.9	101	-	l
Water		cooling system	90	94	-	l	90	98	-	l
Evaporator	300									
Refrigerant		6.5 ⁵	4.9	101 ⁷	-	l	4.9	41	-	v ⁸
Water (load)		building system	60	55	-	l	60	45	-	l
Absorber	360									
Solution		6.0 ⁵	62.7	110	0.62	l	67.6	97	0.57	l
Refrigerant		6.0	4.9	41	-	v	90	abs'd.	-	-
Water		cooling system	90	85	-	l	-	94	-	l
Low Temp Recuperator	116									
St Solution		10	62.7	194	0.62	l	62.7	110	0.62	l
Wk Solution		850	67.6	97	0.57	l	67.6	167	0.57	l
High Temp Recuperator	185									
Intermediate Solution		725	64.9	298 ⁹	0.60	l	64.9	177	0.60	l
Wk Solution		800	67.6	167	0.57	l	67.6	277	0.57	l

1) Gpm for water, pounds per min for brines and refrigerant.

2) Brine concentration, pounds salt per pound brine.

3) 'v' is vapor, 'l' is liquid.

4) 300 mBtu/hr gas input = 225 to solution + 9 jacket loss + 66 flue loss.

5) Vapor pressure.

6) Sub-cooled.

7) Superheated.

8) Refr. Flow is combined flash
loss and load vaporized.

9) 7° drop due to flash in separator.