CONTINUED RESEARCH ON DIRECT CONTACT
HEAT EXCHANGERS - EFFECTS OF CRYSTALLIZATION

- A Final Report

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INTRODUCTION

Direct contact heat exchangers were first proposed by Jacobs and Boehm⁽¹⁾ for extracting heat from geothermal brines in 1973. The recommended use was to overcome perceived difficulties with conventional heat exchangers in binary cycles due to either scaling or fouling (due to the heavy mineral content probable in higher temperature fluids). In addition to being less susceptible to these problems, direct contact heat exchangers can operate with smaller temperature differences between the two fluids. Further, proper design can provide near counter-current operation.

In a review of a variety of direct contact apparatus for use as heat exchangers, published in 1977⁽²⁾, it was pointed out that devices with internals, such as sieve tray towers, Lightnin towers, etc., were more likely to be adversely affected by fouling or scale buildup than were either baffle tray or spray column type towers. This is due to the fact that scale or precipitates could alter the bubble orifices in sieve tray towers and corrosion could effect rotating equipment or seals in devices like the Lightnin tower. Thus, much of the early attention was directed toward spray columns. At East Mesa, California, a 500 KWe direct contact binary geothermal power plant was constructed and successfully operated by Barber-Nichols Engineering using a spray column type direct contactor operating as a combined preheater-boiler.⁽³⁾

The early test sites for direct contact geothermal power systems included the Raft River, Idaho, site operated by EG&G for the U.S. Department of Energy as well as the East Mesa site. Both of these

sites were low temperature (under 150-160°C), low salinity liquid resources. The composition of the East Mesa brine is shown in Table 1 together with that of a high salinity resource near the Salton Sea, California, and a moderate salinity resource at Cerro Prieto in Mexico. The Raft River site produced brine of such low salinity and mineral content that it could easily be used for agriculture! Such sites as Raft River or East Mesa are of only marginal economic applicability even using direct contact binary cycles with their low pinch point. It was felt, however, that they offered one of the severest conditions for direct contact equipment from an economic standpoint. The fact that the brines at these resources were of low scaling potential led to a reconsideration of direct contactors with internals. Thus, EG&G carried out a wide range of experiments with a sieve tray tower, first at Raft River and currently at East Mesa.⁽⁴⁾ This latter work included studies conducted at working fluid critical conditions as well as with working fluid mixtures. The studies, to date, indicate that direct contact heat exchangers can be designed which offer both improved performance and potential economic advantages over conventional heat exchangers in binary power systems when used with relatively clean geothermal brines. This is true for both spray columns and sieve tray columns. Field data is missing for other direct contactor configurations. Despite these favorable conclusions, it is noted that further work is necessary to totally optimize column design, even for spray and sieve tray columns.

As shown in Table 1, high temperature geothermal brines may contain much higher levels of total dissolved solids than the lower temperature brines from East Mesa and Raft River. The Salton Sea

TABLE 1

Composition of Typical Brines* in mg/l

	<u>Salton Sea</u>	<u>Cerro Prieto</u>	<u>East Mesa</u>
Na	56,700	7,800	730
K	11,700	1,660	85
Ca	3,600	545	9
Cl	144,300	14,400	510
Fe	275	NR	0.1
Mn	680	NR	ND
Ag	2	NR	NR
Zn	220	NR	0.1
Pb	38	NR	0.6
Cu	1	NR	NR
Ba	155	NR	0.2
Si	200	400	128
Mg	125	1.4	ND
NH ₄	380	NR	2
SO ₄	100	17	183
HCO ₃	50	50	845
Li	NR	18	0.6
B	NR	18	4
T.D.S.	218,524	24,909	2,498
pH	5.7	8.2	6.3
Downhole			
Temp.	246°C (475°F)	199°C (390°F)	183°C (345°F)

*Proceedings: Seventh Annual Geothermal Conference and Workshop, Paper RP 1525-2, Reference No. 5.

brine is typical of a high temperature high salinity brine. When using such a brine, it is desirable to inhibit brine flashing in the production wells, as flashing may lead to precipitation of minerals within the well casings or in the surrounding field resulting in decreased flow and eventual shutdown of the well or even the field. This, of course, has led to work on the development of downhole pumps in order to insure the production at the well head of a compressed liquid. Although considerable work has been accomplished, reliable downhole pumps are yet to be realized.

Once the brine reaches the surface, the extraction of energy can be accomplished in several ways. For brines above 180°C, a simple flash by throttling to a lower pressure can produce steam to be expanded through a turbine, or multiple flashes can be used with multi-port turbines allowing for steam inlet at different pressures. Typically, throttling generation of steam can be practically achieved to temperatures of 115°C (169 KPa). References 5 and 6 discuss the possibility of a double flash for a brine characterized by the Salton Sea brine of Table 1. They propose a first flash to 1 MPa followed by a second to 169 KPa. These conditions are, of course, the conditions of the dual flash test facility constructed by San Diego Gas and Electric and Magma Power at the Salton Sea.⁽⁷⁾ The dual flash facility encountered major scaling problems in the second stage flash tank, and the atmospheric flash tank and the associated piping and valves after only 1000 hours of operation. Also, supersaturated silica and suspended solids in the spent brine have caused scaling and plugging of the injection pump casing and injection well casing.

Scaling also took place in the high pressure flash tank; however, the type of scale differed considerably. Ahead of the first stage flash tank a black scale formed which contained heavy metal sulfides. Beyond the first stage flash vessel, the galena rich scale became less abundant and silica scaling became more pronounced.

Several methods were considered to reduce or eliminate the scaling. These included chemical treatment with phosphonate type chemicals, hydroxyethylcellulose and acid. Silica precipitation is both pH dependent and temperature dependent. Thus, acid treatment has been shown to inhibit scaling to a greater degree than any of the other additives.^(7,8,9) Reference 7 reports a predicted 11¢/KWhr increase in cost for a geothermal power plant using acid treatment; thus, other techniques for scale reduction were explored for flash type systems.

Reference 7 reports that a reactor-clarifier-filter system can be effective in treating the reinjection brine. Details of such a system are described by Dahlstrom et al.⁽⁹⁾ The system can reduce dissolved silica to less than 170 ppm and suspended solids to 4-10 ppm. These levels appear acceptable for reinjection fluids.

The successful use of a pilot reactor-clarifier led Magma power to the idea of using the same technique to reduce scale formation in the flash vessels. The possible use of a crystallizer as a flash vessel was first discussed in 1978⁽⁷⁾. This led to a variety of designs.^(5,6,7,8) All of the designs proved effective with the design discussed in References 5 and 6 holding promise of being most effective.

The effective use of crystallizers in the design of flash type geothermal power plants for use with high temperature, high salinity brines led the U.S. Department of Energy to question whether crystallizer schemes could be effectively used in binary geothermal power plants and more specifically direct contact geothermal power plants. This question led to the funding of the present preliminary study.

The present study was to include four technical tasks. They are as follows:

Task 1 - Conduct an industry and literature search for models that predict potential scale formation.

Task 2 - Based on the results of the search, select and modify, if necessary, a model for estimating potential scale formation within direct contact binary power plants and components. Using this model, combined with existing methods for calculating direct contact heat exchange, develop a procedure for application to direct contact binary power plant design for predicting:

- (1) the precipitation and particle-growth behavior from different dissolved species (Note: industry involvement, particularly Bechtel, should be solicited), and
- (2) heat transfer performance.

Task 3 - Evaluate the circumstances under which direct contact internals can be used without adverse scaling problems.

Task 4 - Investigate schemes of utilizing a combined heat exchanger-crystallizer to eliminate adverse scaling problems. Industry should be consulted regarding what has been done in this area.

Of the four tasks proposed for investigation enumerated above, only Task 1 was fully completed. The result of this task indicated that the theoretical models for predicting not only homogeneous nucleation, but also heterogeneous or secondary nucleation are suspect. For example, Reference 18 on pages 102-103, indicates that the theory for homogeneous nucleation of simple binary systems "predicts nucleation only at extremely high supersaturations, a phenomenon not observed in most inorganic crystallization systems." It has been observed, they go on to say, "that in clean systems, where dust and extraneous material have been meticulously removed, that nucleation does not take place even at high supersaturations." In addition, the theoretical models require knowledge of many quantities that are unknown for many systems of solvents and solutes let alone complicated mixtures such as geothermal brines.

Secondary nucleation refers to nucleation or growth of crystals from supersaturated solutions on other surfaces. It is also referred to as heterogeneous nucleation. It is this type of nucleation that occurs in common crystallizers, on the walls or surfaces of pipes, vessels, etc. Heterogeneous nucleation models are presented basically in the form of a Arrhenius type relationship, as are the previously mentioned homogeneous nucleation models. The latter types of nucleation models depend upon the existence of nucleation sites and,

thus, surface area. Although simple models have been formulated, they are again highly suspect. Reference 19 indicates $\pm 100\%$ accuracy is the current state of the art unless specific solutions are evaluated. Similar lack of data was confirmed by Van der Mast at Bechtel and is missing in their work. (5,6)

Because of the lack of adequate models or data to predict nucleation and then crystal growth, Tasks 2,3 and 4 could not be accomplished in total. A conservative estimation of potential hazards of scaling could be predicted, however, by assuming that equilibrium thermodynamics must exist. In such models precipitation would occur as the brine reaches saturation conditions. Such a model would preclude the use of direct contactors with internals as these internals would provide additional nucleation sites for heterogeneous precipitation and nucleation. Thus, in this final report, attention has been fixed on the use of spray columns.

CAUSES OF SCALE

In the reservoir of a geothermal field, the brine is in equilibrium with the rock formation. Minerals go into solution resulting in a chemical equilibrium state for the brine at its temperature and pressure. Production of the reservoir disturbs this equilibrium. As the pressure is reduced, dissolved gases may be released. For example, carbon dioxide exists in equilibrium with soluble bicarbonate:

As CO_2 is released, the equilibrium shifts to an insoluble carbonate which can react with calcium to form the carbonate scale that can plug the well casing or downstream valves.

The precipitation of binary metal compounds is more complex,⁽⁵⁾ involving several sequential reactions:

Thus, it is seen that the reduction of pressure and release of CO_2 can lead to the precipitation of CaCO_3 , PbS and Fe_3O_4 , for example. The release of CO_2 can also change the pH level, making it less acidic. This can lead to supersaturation of other materials including other metal salts and silica.

The use of a direct contact heat exchanger with a boiling section for the working fluid will undoubtedly lead to the release of some CO_2 as well as significant cooling of the brine. The effect would be not unlike that occurring in a first stage flash process. It is, thus, likely that precipitation of metal sulfides will occur as was noted in Reference 7.

The precipitation of silica is, however, predominately a physical process. As shown in Figure 1, which is reproduced from Reference 7, the solubility of either quartz or amorphous silica decreases with decreasing pure water temperature for temperatures below 300°C . From crystallizer tests with Salton Sea geothermal brines, a similar behavior is seen. Since the crystallizer was in all cases a flashing device, it is not clear that this is strictly a temperature effect as one might surmise from the curve. The pH level is increasing as the temperature decreases since a lower pressure flash was obtained for each lower temperature.

References 10-12 indicate that the solubility of amorphous silica is from 25-60% lower in sodium nitrate solutions than in pure water. Reference 11 indicates nearly the same level of solubility in NaCl as NaNO_3 at 25°C ; thus, the effect noted in Figure 1 for the crystallizer is probably primarily the effect of salt concentration and temperature.

Since, in a direct contact binary power plant, it is not necessary to achieve large pressure drops from the well head conditions, it is not necessary to have liberated large amounts of carbon dioxide gas. In fact, depending upon the conditions chosen for the working fluid exiting the direct contactor, little or no water vapor may be produced. Thus, the chemistry, pH and concentration need not have changed except as effected by the decreasing temperature of the brine as it flows down the direct contactor. Therefore, for direct contact binary cycles, it is probable that the only scale problem is due to amorphous silica precipitation. Of course, downstream of the direct contactor precipitation of silica or other materials could be as bad as in the flash system. This is due to the possible desire to remove any of the working fluid that may have gone into solution in the spent brine and needs for clean up prior to reinjection in the field.

If a given heat exchange is desired in a direct contactor for a given binary system, working fluid and brine, the brine will be subjected to essentially the same temperature profile along the length of the exchanger. The length of the heat exchanger and the residence time of the brine will change with the type of direct contactor chosen. A direct contactor with internals could provide a higher heat transfer coefficient than a spray column, thereby, leading to a shorter residence time for the brine in the direct contactor. It will, however, offer considerably greater surface area from its walls and internals, promoting an increase in heterogeneous nucleation sites. A spray column direct contactor offers only the walls of the direct contactor on which scale can form in addition to the working fluid

injectors. It does offer, as do DCHX's with internals, the possibility of heterogeneous nucleations sites on the working fluid drops. No data exists to ascertain whether this phenomenon is a viable means of promoting crystallization. (19) If it is not, then only homogeneous nucleation is left for inducing crystallization in a spray column type direct contactor. Homogeneous nucleation, while it cannot be easily modeled, is known to be hard to induce. (18,19) Typically, it only occurs at very high levels of supersaturation, and it takes a long time to produce any significant degree of precipitation. (Typically several hundred seconds (19)). A long time is considered here to be hundreds of seconds or several minutes. Thus, unless there are means of causing heterogeneous nucleation, such as internals, it is likely that that scaling of silica will only take place on the containment walls of direct contactors. Direct contactors can, thus, be used to cool a brine to a supersaturated state leaving the majority of precipitation to occur downstream of the heat transfer zone. It should, thus, be possible to include a variety of classical crystallizer schemes downstream, in a brine sense, of the working fluid distributor.

POTENTIAL DIRECT CONTACT BINARY POWER PLANT DESIGNS

A wide range of possible binary cycles can be proposed for use with brines typified by the Salton Sea brine of Table 1. Figure 2 shows a supercritical cycle with the brine entering as a compressed liquid. The compressed liquid state could be achieved by a downhole pump if such were available and needed, or by a well head pump. The pressure in the direct contactor is chosen to optimize the cycle for the working fluid chosen. The high salinity of the brine, 218,524 ppm, keeps the solubility of the working fluid, let's say isobutane for example, to a minimum. If the brine leaves the direct contactor, at temperatures of 50-60°C, negligible precipitation of silica may result. Our schematic, after the direct contactor, shows energy recovery and pressure reduction by expansion through a Pelton wheel. By expansion to atmospheric pressure, working fluid loss can be reduced to 1 ppm by weight in the brine which would then go to a reactor-clarifier such as described in Reference 9. Although the brine could exceed supersaturation of silica in the direct contactor, the lack of release of CO₂ would keep the pH at the 5.7 level. This would allow for somewhat higher solubility of amorphous silica than indicated in Figure 1; however, the brine could be strongly supersaturated if the amorphous silica - Salton Sea curve is applicable (100% supersaturation). Although not shown in Figure 2, a crystallizer could be included before the Pelton wheel, if needed. Such a crystallizer would require the injection and recirculation of seed material similar to Reference 9. No pressure drop would occur in this system, but only across the Pelton wheel.

A combined flash steam, binary cycle is shown in Figure 3. In this cycle, the brine is flashed from 246°C to 180°C. This is equivalent to the conditions of the first stage flash of the San Diego Gas and Electric Company, Magma, Inc., dual flash system at the Salton Sea Niland facility. Low levels of precipitation of heavy metal sulfides and CaCO_3 will occur in the flash tank eliminating them from the direct contactor. Instead of the second flash, the brine exiting the first flash tank is compressed to 2.2 MPa. It then flows into the direct contactor where temperature and pressure conditions exist similar to those at the East Mesa DCHX facility. The brine exit temperature of approximately 60°C would again minimize precipitation of silica, even considering the process to be at equilibrium. Brine at 2.2 MPa can, again, be expanded through a Pelton wheel to extract part of the energy that was necessary to compress the brine from 150 KPa to 3.4 MPa as well as recover the isobutane down to 1 ppm. The use of a crystallizer stage could be introduced prior to the Pelton wheel in the same manner as for the supercritical cycle discussed above. Again, the spent brine leaving the Pelton wheel would go to a reactor-clarifier prior to being reinjected into the field.

A third possible cycle that might be contemplated is a subcritical, isobutane binary cycle as shown in Figure 4. In this cycle, brine straight from the well head is used to heat the working fluid and vaporize it. Some water vapor will leave the direct contactor with the isobutane vapor at a ratio of 0.0124Kg/KgIC₄. This is equivalent to 2.3% vaporization of the H₂O in the brine. It compares to 14.66% vaporized in the the first stage flash. Thus, although there will be some precipitation of heavy metals and CaCO_3 ,

it will be considerably less than even in a first stage flash. An optimum cycle for isobutane would produce isobutane vapor in a direct contactor at a temperature of 127°C and 3.22 MKPa. The brine would leave the direct contactor at 42°C. Whether precipitation of silica or other compounds would occur within the direct contactor would depend upon, not only the amount of potential precipitant present and the solubility limits, such as given in Figure 1 for silica, but also the time duration of the supersaturated brine at temperature within the direct contactor, as noted earlier. Figure 1 would indicate that the solubility limit of silica in pure water would just be reached at the exit of the direct contactor, while the silica-crystallizer curve would indicate the solubility limit in the Salton Sea brine could be reached at the 100°C temperature level.

For a spray column designed for the subcritical cycle shown in Figure 4, the length of the preheater would be nearly six meters in length. This length was calculated using the design procedures described in References 13 and 14. The saturation condition for Salton Sea brine silica (see Figure 1) would be reached at a distance of 3.5 meters above the isobutane distributor/injector. If the pure water amorphous silica curve more closely represents the solubility, then saturation would be reached at only 1.4 meters above the distributor. As the brine mean velocity in the column would be at about 7 mm/sec between the distributor and these two levels of potential saturation conditions, the residence time of saturated to supersaturated brine in the column would be between approximately 500 seconds and 200 seconds. The former time is similar to that necessary for 58% relief of supersaturation in the second stage crystallizer--

flash units described in Reference 5. The latter time is equivalent to less than 15% relief. If these types of relief were achieved, the amount of silica precipitated could be between 86 Kg/hr and 6 Kg/hr, respectively, in a 50 MW_e direct contactor for the cycle of Figure 4.

Silica precipitation of the amounts described above could only occur for the case of heterogeneous nucleation. In other words, by the introduction of a highly efficient nucleation site generator within the direct contactor. This would require introduction of silica particles into the column at a point near the initial saturation level of a size and density that would allow them to fall toward the isobutane distributor, but to be collected prior to reaching it and to be directed around it. Circulation of particles back to the particle injector level would also need to be accomplished. Particle buildup on the isobutane injector would have to be avoided. Buildup of particles or scale at the bottom of the column could be tolerated and a particle settler and removal scheme could possibly be developed.

It is possible, however, that little or no silica precipitation could occur in the direct contactor other than on the vessel walls. If the solubility of silica followed the amorphous silica pure water curve, the 200 second residence time is probably too short for homogeneous nucleations to have much effect. (19) However, the 500 second residence time is sufficiently long that some precipitation may occur. In order to evaluate whether precipitation would occur in a spray column, it is necessary to have experimental data for homogeneous nucleation and for heterogeneous nucleation without as

well as with accompanying pressure reductions. Particle growth rate information is also needed. It is also necessary to have reliable solubility data for potential precipitates for the brine chemistry of interest. The latter is needed for even the highly conservative assumption of equilibrium. The nucleation data is needed in order to design to avoid precipitation, or to plan for it with any acceptable degree of confidence.

The use of internals in a direct contactor is possible if they are used in locations where the saturation limit of possible precipitating solutes has not been reached. They could also be used if they are of a material that would not provide nucleation sites. Such material would have to be hydrophobic. The primary problem with hydrophobic surfaces is that they are probably oleophilic and, thus, not acceptable for sieve trays, thus, eliminating the use of sieve tray columns.

CONCLUSIONS

The use of a variety of direct contact geothermal binary cycles can offer advantages in terms of fouling or scaling over flash systems for high temperature, high dissolved solid brines. The primary precipitant in direct contact systems appears to be amorphous silica since its solubility is primarily dependent on brine temperature.

The presence of salt in the brine will normally decrease the temperature at which the solubility limit or saturation condition is reached based upon limited data available in the literature; thus, temperatures as high as 100°C could initiate silica precipitation. Such temperatures are easily reached in the preheater section of a direct contactor as temperatures as low as 42°C are reached in optimum cycles. However, available solubility data for amorphous silica in pure water indicate that temperatures as low as 60°C can be reached prior to the saturation limit for silica. For this solubility limit, it should be possible to avoid precipitation in a spray column above the working fluid injector. This would allow the use of a standard design crystallizer immediately following the direct contactor to precipitate out the silica, thereby avoiding fouling of downstream equipment and the reinjection well.

In order to ascertain the best overall system design, it is mandatory that more reliable data be obtained on equilibrium solubility curves for geothermal brines. It is also necessary to establish an experimental data base for both heterogeneous and homogeneous nucleation of precipitation for the minerals in geothermal brines. Precipitation kinetics for particle growth rate are also necessary.

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TABLE 2

Operating Conditions for System of Figure 4.

Net Power - 46 MW
Working Fluid - Isobutane
Mass Flow Rate of Isobutane - 2.39×10^6 Kg/hr
Mass Flow Rate of Brine - 1.305×10^6 Kg/hr
Mass Fraction of Steam in Isobutane Vapor - 0.012
Fluid Lost Hr - 1.31. Kg/hr IC_4
Turbine Exit Quality - 0.9961
Turbine Exit Water Quality - 0.6765
Turbine Speed - 5027.8 PRM
Turbine Diameter - 1.305 meters
Utilization Factor - 0.180
Thermodynamic Figure of Merit - 0.140
Direct Contactor Dimensions Cross-Sectional Area - $60.8m^2$ *
Boiler Height - 0.2m
Preheater Height - 5.6m
Droplet Diameter - 3.5mm

*Could be composed of several columns in parallel.

Figure 1 Solubility of SiO₂ from Reference 7

Figure 2 Super Critical IC_4 Binary

Geothermal Power Plant for Use With High Temperature, High Salinity Brine

Figure 3 Combined Flash Binary IC_4 Geothermal

Power Plant for Use With High Temperature,
High Salinity Brine

Figure 4 Subcritical Binary IC₄ Geothermal

Power Plant for Use with High Temperature,
High Salinity Brine