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IMPROVING THE INJECTABILITY OF HIGH SALINITY BRINES FOR DISPOSAL OR WATERFLOODING OPERATIONS¹

Ellen Raber², Robert E. Thompson³, and Floyd H. Smith⁴

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ABSTRACT

This work is part of a study conducted by Lawrence Livermore National Laboratory (LLNL) to improve the performance of brine injection wells at Gulf Coast Strategic Petroleum Reserve Sites. Our involvement established that granular media filtration, when used with proper chemical pretreatments, provides an effective and economical method for removing particulates from hypersaline brines. This treatment allows for the injection of 200,000 B/D with significantly increased well half-lives of 30 years.

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INTRODUCTION

The ability to inject brines into permeable geologic formations is important in a variety of industrial applications: (1) for the disposal of leached salt dome brines utilized for chemical extractions and/or oil storage, (2) as a formation pretreatment in waterflooding operations for enhanced oil recovery, (3) as a means of disposal for spent brine effluents from geothermal and/or geopressured resources, and (4) as a means of reservoir pressure maintenance. In all of these applications, it is not only important to measure the clarity of the brine by conventional wastewater schemes, but it is essential to produce a high quality effluent which is in chemical equilibrium with the injection formation. Otherwise, flow through the geologic media will be inhibited by precipitation and plugging, resulting in formation damage and well impairment.

We have recently been involved in evaluating various filtration methods as possible means of clarifying and improving the injectability of hypersaline salt dome brines (30 weight percent NaCl). It was our intent to establish which combination of filtration method and chemical pretreatment would yield the highest quality effluent for reinjection. This work involved extensive field tests at three Strategic Petroleum Reserve (SPR) sites (Bryan Mound in Texas and West Hackberry and Bayou Choctau in Louisiana) where injection flowrates averaged 10^5 B/D and wells were accepting brine at 50 percent or less of their initial design rates.

The state-of-the-art convention is to use granular media filtration to clarify effluents prior to subsurface dispersal. Most applications involve treatment of moderate to low salinity waters with total dissolved solids of less than 100,000 ppm.¹⁻² However, application of granular media filtration as a means of clarifying hypersaline brines (>200,000 ppm) for injection was successfully demonstrated at the Salton Sea Geothermal Field in southern California.³ Since granular media filtration depends on adsorption, the strong electrolytic effects of hypersaline brines on colloid charge stability and sorption will influence the performance of granular media filtration and the selection of prefiltration chemical aids.

TEST OBJECTIVES AND METHODOLOGY

The objectives of this paper will be to present the methodology and requirements necessary to remove colloidal solids and produce an effluent which will not precipitate in the geologic formation and impair injection well longevity. Figure 1 shows the methodology employed to evaluate the injectability of a fluid. This involves an initial field evaluation which includes the following techniques.

Incubation tests were run to assess time-dependent changes in particulate concentration and to determine the potential for post-precipitation and possible wellbore impairment. Brine is sealed in airtight containers and incubated at the injection temperature for periods of up to one month.

Compatibility tests of injection fluid with reservoir or formation connate waters are sometimes necessary. These tests are similar to the incubation tests previously described and again determine the potential for postprecipitation due to geochemical disequilibrium.

Core flooding tests are utilized to assess the geochemical equilibrium of the fluid with the geologic formation. These tests are not necessary when the injection formation is unconsolidated as is the case with this particular study.

Filter membrane tests were conducted to assess the plugging of the formation due to suspended particulates. The use of membrane filtration tests as an aid in determining water quality requirements for injection has been well described in the literature. LLNL has developed field apparatus to measure the decline in flowrate versus the total cumulative volume at constant differential pressure, allowing an evaluation of effluent injectability.⁴

Initial field evaluations showed that direct injection without processing was not feasible due to high suspended solids and short injection well half-life. Injection well half-life is defined as the time required for the injection rate to fall to one-half of its initial rate at constant pressure. LLNL uses a modified version of the method described by Barkman and Davidson⁵ to predict the rate at which suspended solids plug injection wells.⁶ This incorporates a method described by Champlin et al.⁷ to estimate the average pore diameter of the injection formation and chose a membrane filter pore size which better simulates the nature of the injection zone.

The injection formations at the SPR sites were unconsolidated Miocene sands with a 30% porosity and 0.8-5.0-darcy permeability. Results from initial tests showed untreated effluents allowed unacceptable well half-lives of only 0.25 years.⁸ Therefore, some type of pre-injection process development was required to improve effluent quality. The subsequent methodology we employed in our field tests to determine the clarification and processing requirements can be seen in Fig. 2.

FIELD TEST PROCEDURES

Field test procedures followed the general methodology described in Fig. 2. Based upon the concentration of suspended solids and chemical composition of the brine, the main emphasis was placed on evaluating downflow granular media (combinations of coal, garnet and/or sand) filters. The test procedure at each of the three sites was divided into four parts.

1. Pilot tests of direct filtration without chemical coagulants were performed with downflow granular media filters, ultrafilters, and cartridge filters.
2. Inorganic salts and polymers were evaluated as coagulants/flocculants by a combination of jar testing and bench-scale filtration to identify the chemicals that could significantly improve granular-media filtrations.

3. The performance characteristics of downflow granular media filters were evaluated with brines that were pretreated with chemicals that were found in jar tests to be effective in coagulating particulates contained in these hypersaline brines.

Six combinations of filter media were evaluated at the three sites. The construction of these 4-in diameter subpilot filters is given in Table 1. Listed media sizes were determined by standard sieve analyses. A constant feed rate of 8 gal/min/ft² was usually maintained for all granular-media filter runs, and filters were restored to normal conditions by backwashing at a rate of 16 gal/min/ft² for five to eight minutes. Many full-scale granular-media filtration plants have been successfully designed utilizing test data from similar subpilot filters.⁹ In addition to the granular-media filters, we also evaluated hollow fiber ultrafilters and disposable cartridge filters for use at SPR sites.

RESULTS FROM FIELD TESTS

Filter performance was evaluated with regard to (1) pressure loss versus time (headloss), (2) effluent quality (turbidity), (3) length of filter cycle, (4) particle size distribution, and (5) injectability with respect to the permeability/porosity of the injection formation. Due to various degrees of contamination from oil, minor differences in brine chemistry, and variable brine dilutions, each site must be evaluated independently.

1. Evaluation of Direct Filtration Without Chemical Pretreatments

Ultrafiltration produced a highly injectable quality brine effluent and good filter cycle times without the necessity of chemical pretreatment (turbidity <0.12 NTU), alleviating problems associated with chemical additives and changing brine conditions. However, more testing is necessary before a definitive statement as to long-term effectiveness can be made. There is no industrial experience with ultrafilters having capacities of 150,000 to 200,000 B/D.

Disposable cartridge filters ($1\ \mu$ pore size) effectively reduced suspended solids without the use of chemical aids. However, they plug too rapidly and frequent renewal would not be practical for the treatment of large quantities of brine. Calculations estimate that any site would need 360,000 cartridges per day.

Granular media direct filtration with no chemical treatment usually produced unacceptable quality effluent for injection although, occasionally, an acceptable quality effluent was produced. This suggests that the brine is sometimes at an electrolytic state in which the diffuse layer of ions around the particle surface is sufficiently compressed, allowing some coagulation without the use of chemical additives. Figure 3 shows the head loss versus time at the West Hackberry SPR site. Increase in headloss is directly related to solids loading and short filter run times. Only dual and triple media filters had acceptable headloss rates. Since the effluent quality was only sometimes acceptable, further evaluations were done with chemical pretreatments.

2. Evaluation of Chemical Pretreatments

One of the most important aspects of particulate removal is the use of coagulants/flocculants. These chemicals cause destabilization of the particle surface charge allowing particle agglomeration which enhances removal by filtration. Although inorganic and organic coagulants are used extensively in the wastewater industry, their effectiveness in hypersaline brines is not well established.

Over fifty inorganic salts and polymers were evaluated by a combination of jar testing and bench-scale filtration techniques. These experiments included tests on stagnant recirculated brine and cavern leach brines at various concentrations. Samples were mixed at ambient temperature for two minutes at a constant speed of 100 rpm and then at 20 rpm for ten minutes. This procedure permitted particle destabilization and flocculation. Turbidity measurements (Hach model 2100A) were made on the supernatant, which was

prefiltered by gravity through Whatman #2 paper. This technique has been shown to produce effluent quality similar to that produced by the granular media filters.⁹ The results of these tests can be seen in Table 2.

Jar tests showed initially that alum and moderately charged high-molecular-weight anionic polymers were the most effective with regard to treatment of hypersaline brines. Average turbidities were lowered from 10 to 0.20 NTU after addition of these chemicals. The highly charged anionic polymers often caused dispersion of suspended particulates. Cationic polymers were not found to be very effective. In addition, long-chained nonionic polymers sometimes coagulated particulates, suggesting that a high degree of particle coagulation must already exist and further filter enhancement can be provided by a bridging agent.

It should be noted that anionic polymers have also been found to be effective coagulants in hypersaline geothermal brine.³ This is in contradiction to the cationic polymers that are usually found to be the effective coagulants in normal low-salinity waste-waters. Colloids in aqueous solutions have a double layer composed of water molecules and ions counter in charge to the particle surface charge. This layer is very diffuse and loosely bound in low salinity (low electrolyte) solutions. Coagulation releases the bond water layer and allows the particles to agglomerate. Since most colloids in aqueous solutions have a negative surface charge, it usually takes a positive cationic polymer to coagulate. High-salinity (high electrolyte) solutions have an extremely compressed, tightly bound double layer. Therefore, we speculate that the surface charge is positive and anionic polymers are necessary to cause coagulation/ agglomeration.

3. Granular Media Pilot Tests with Chemical Pretreatments

Granular media filtration was an effective means for hypersaline brine clarification with the correct chemical pretreatment. Field tests show that both dual and triple media configurations produced a high quality injectable effluent (turbidity < 0.20 NTU) with acceptable headloss rates and filter cycle

times. These media constructions with pretreatment were as effective as ultrafiltration, although more sensitive to changing brine conditions. A comparison of improved effluent particle size distributions can be seen in Fig. 4.

Jar tests and bench scale filtration tests were used to provide data on the optimum chemical dosages for coagulation. However, these procedures must be followed by tests to assess filter performance with respect to various media design.¹⁰ In the pilot filter tests, these prescreened chemicals were injected into the influent stream. While jar testing had indicated that several chemicals would coagulate particulates, only alum or the high-molecular-weight anionic polymers produced acceptable quality effluents when actually applied to multi-media filters. Long-chained nonionic polymers used in conjunction with alum could sometimes increase floc shear strength and reduce filter penetration.

It can also be concluded that a prescribed chemical pretreatment could be made ineffective under changing brine conditions. Preliminary evidence suggests dilution of brines affects polymer coagulation performance, but further studies are needed. At the West Hackberry and Bryan Mound sites, coagulation tendencies of anionic and nonionic polymers seemed to be inhibited by oil contamination. Polymers alone were effective only at Bayou Choctaw where oil contamination was not a problem.

SUMMARY OF FIELD RESULTS

A summary of the most effective systems for brine clarification at each of the three sites is presented in Table 3. The specific pilot tests and other evaluations conducted at each site will not be described in this paper. However, bench scale tests at all sites demonstrated that sedimentation and chemical pretreatment with settling do not yield an acceptable quality brine effluent for injection.

Both ultrafiltration and multi-media filtration with the proper chemical pretreatment were found to be extremely effective. Headloss and length of filter cycle (Fig. 3) were acceptable for both dual and triple media filters. Particle size distribution (Fig. 4) improved such that the majority of particles were below the $1\ \mu$ detection limit.

Figures 5 and 6 show further comparisons of improved effluent qualities and improved injectabilities for the recommended granular media filtration schemes and ultrafiltration with the original raw brine. Calculations from Fig. 6 show untreated effluents allowed well half-lives of only 0.25 years, while pretreatment of brine by the recommended methods increased injection well half-life in excess of 30 years.⁸

PROBLEMS ASSOCIATED WITH CHEMICAL PRETREATMENT

Postprecipitation potential

Incubation tests were run on chemically clarified effluents at 35°C for periods ranging from 1 to 24 hours. Samples were filtered through $1.0\ \mu$ membrane filters to determine the tendency for postprecipitation. Discussions with leading chemical companies suggested that problems of downhole postprecipitation due to chemical additives were especially likely in the presence of either Fe^{2+} (as little as 0.2 ppm) or Ca^{2+} (200 ppm).

Bryan Mound brine, which contained the highest concentrations of these ions, was used for these experiments. The test results show that postprecipitation is 0.1 ppm in the presence of excess alum (at 3 ppm), excess alum + Cyfloc 4500 (at 10 ppm alum + 0.2 ppm nonionic polymer), and excess anionic polymer Visco 3340 (at 3 ppm). If chemicals are employed in filtration operations, incubation tests must be done on the filtered effluents to determine the potential for downhole postprecipitation.

Residual Polymer

Field observations indicated that residual polymers can contribute to the plugging of membrane filters. Follow-up laboratory studies were conducted with prefiltered brine, and additional injectability tests were performed. A relationship was observed between residual polymer concentration and loss of filter permeability. Additional laboratory work is being conducted to unequivocally establish the plugging mechanism. Such studies are desirable since injection wells could be impaired by the deposition/adsorption of residual polymers or by the coagulation of trace particulates in post-filtered effluents.

BRINE TREATMENT SYSTEMS DESIGN

The downflow granular filter approach offers flexibility because it can operate effectively with or without chemical addition to the brine. Any size or configuration of pressure filters may be utilized. However, to provide a filter system that is versatile to install and use as well as easy to ship, a standard module is proposed consisting of a support frame 10 feet wide and 20 feet long. The design schematic for this system can be seen in Fig. 7. Each filter tank is connected to the two main headers running the length of the assembly and can be individually isolated from both headers by operating manual valves. The upper header is used for raw water transfer to the top surface of the filter media during filtering mode of operation and for removal of backwash wastewater during backwashing mode. Skid assemblies are manifolded together for backwashing purposes. Multiple filters can be manifolded together as required to meet the desired flow rates at each injection site. Automatic controls for this system consist of two open-closed valves which are used to divert the normal flow for each filter from filtering mode to backwashing mode, and one modulating final effluent valve which is used to control filter effluent rate. Backwashing is initiated by high headloss across the filter or manually as desired by the operator. The estimated cost of this approach, including the main piping system required to connect the necessary number of filters together to obtain a properly sized

treatment plant, is \$840,000. Manufacturing time required for this approach appears to be approximately 20 to 24 weeks for a nominal plant capacity of 250,000 B/D.

The ultrafiltration approach provides the best potential quality of all the systems described, however, as stated earlier, there exists no industrial experience for capacities of 200,000 B/D. A pilot unit was field tested at both Bayou Choctaw and Bryan Mound. The single ultrafiltration cartridge system that was tested can provide an effluent with suspended particles of less than $0.1\text{ }\mu\text{m}$. This system is easy to operate as no chemicals are required for optimum effluent qualities. This type of filter process is strictly a mechanical separation of clean water from dirty water through the utilization of pressure to force a varied percentage of the raw water through a semipermeable membrane. The reject water that does not pass through the membrane is returned to the brine pond. Periodic backflushing and chemical cleaning of the membranes is required and is accomplished in much the same manner as backwashing the granular filters previously described. The estimated cost for this approach, including the main piping system, is \$1,700,000. Manufacturing time required for this approach is approximately 30 weeks for a nominal plant capacity of 250,000 B/D.

CONCLUSIONS

Hypersaline brines can be injected at capacities of 200,000 B/D, resulting in increased well half-lives in excess of 30 years. Processing requirements and assessment of brine injectability can be established utilizing the systematic methodologies presented in this paper. For Strategic Petroleum Reserve Sites, where decline in injection well performance was due mainly to high particulate concentration, both multi-media filtration with chemical pretreatment and ultrafiltration produced a high quality injectable brine effluent. High molecular weight polyacrylamide anionic polymers exhibited the most versatility as a chemical pretreatment, although, alum or alum plus a nonionic polymer was found more effective during periods of oil contamination. A modular downflow granular media filtration system can be

designed so that it is both flexible and economical. Ultrafiltration, although not susceptible to changing brine conditions and easier to operate (requiring no chemicals), has limited industrial experience and is more costly.

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Table 1

CONSTRUCTION OF 4" DIAMETER PILOT FILTERS

Filter	Construction	Sites Tested
Single-media	12" silica sand	West Hackberry
Dual-media	12" garnet 18" anthracite coal	Bayou Choctaw
Dual-media	12" silica sand 18" anthracite coal	Bayou Choctaw and West Hackberry
Triple-media	3" garnet 9" silica sand 18" anthracite coal	Bayou Choctaw, West Hackberry, and Bryan Mound
Ultrafilter	Romacon hollow fiber cartridge; 3 in. with 525-ml volume	Bayou Choctaw and Bryan Mound
Disposable cartridge	A.M.F. Cuno, 1.0 μ cartridge filters	Bayou Choctaw, West Hackberry, and Bryan Mound

Grain Size silica sand = 0.45 - 0.6 mm
garnet = 0.28 - 0.35 mm
anthracite coal = 1.0 - 1.1 mm

Table 2

BENCH SCALE EVALUATION OF COAGULANTS AND FLOCCULANTS

Coagulant/Flocculant	Amount Added ppm	Description	Effect on Turbidity at Indicated Site		
			West Hackberry	Bayou Choctaw	Bryan Mound
<u>Anionics</u>					
Chlorine	1-5	—	N	N	N
Calgon M-570	0.5-3	Medium molecular weight (7 m)	N	—	—
Calgon M-580	0.5-20	Medium molecular weight (7 m)	R	—	—
Calgon M-590	0.5-10	Medium molecular weight (7 m)	N	R	—
Cyanamid-Cypan	1-3	200,000 MW	—	—	D
Cyanamid-CY 292	1-3	400,000 MW	—	—	D
Cyanamid-Cy-Guard 382	0.5-10	10,000 MW	—	—	D
Cyanamid-P-26	0.5-10	150,000 MW	—	—	D
Cyanamid-5300	0.5-5	15 million MW	—	—	R
Magnafloc 834A	0.5-10	High molecular weight (18 m)	—	R	R
Magnafloc 837A	0.5-10	15 million MW	—	R	—
Magnafloc 1820	0.5-10	High molecular weight (18 m)	—	R	—
Visco 741	0.5-5	15-20 million MW	—	—	R
Visco 743	0.5-5	10-15 million MW	—	—	N
Visco 3340	0.1-10	Long-chained, high molecular weight (18-20 m)	E	E	R
<u>Cationics</u>					
Alum	1-300	Inorganic, short-chained, high-charged used with Cyfloc 4500	E	R	E
FeCl ₃	1-50	Inorganic, short-chained	R	—	—
Calgon-Cat Flocc-T	0.5-10	Low molecular weight (0.5 m)	N	N	—
Magnafloc 507C	0.5-3	Low molecular weight, high-charged; used with 3340	N	N	—
Magnafloc 581C	0.5-10	—	N	N	—
Magnafloc 1561C	0.5-10	—	—	N	—
Magnafloc 1563	0.5-10	—	—	N	—
Nalco Vx-740	0.5-5	High molecular weight (7-10 m)	—	—	N
Visco 3317	0.5-3	AlCl ₃ cationic polymer; used with (834A, 1820A, 3340)	N	R	R
Visco 3342	1-20	—	N	—	—
Visco 3347	0.5-10	Alum; cationic polymer	N	N	—
Visco 3349	0.5-10	—	N	N	—
Zimmite 2T68	1-20	—	N	—	—
Zimmite 2T653	0.5-20	—	N	—	—
Tretolite-FR-52	1-20	—	—	—	D
<u>Nonionics</u>					
Cyfloc 4500	0.02-5	Long-chained, high molecular weight (15 m); used with alum, 3317, and 3340	E	N	E
Magnafloc 985N	0.1-10	Long-chained, high molecular weight	R	N	R
Magnafloc 990N	0.5-10	Medium molecular weight (4 m)	N	—	N
Polyethylene Amines	0.5-20	Low molecular weight	N	—	—
Polyglcol P-400	0.5-20	Low molecular weight	N	—	—

N = no change in turbidity

R = reduced turbidity to some degree

E = excellent turbidity reduction

D = dispersed

Table 3
RECOMMENDED GRANULAR MEDIA CLARIFICATION
SYSTEMS FOR SPR SITES

Site Location	Chemical Additive	Concentration, mg/l	Type	Media Construction
West Hackberry	ALUM $\text{Al}_2(\text{SO}_3)_4 \cdot 14\text{H}_2\text{O}$	3	Inorganic Al salt	Dual media (coal, sand) or triple media (coal, sand, garnet)
Bayou Choctaw ^a	Visco 3340	2-4	Anionic polymer	Triple media (coal, sand, garnet)
Bryan Mound ^a	ALUM + Cyfloc 4500	10 + 0.2	Inorganic Al salt + nonionic polymer	Triple media (coal, sand, garnet)

^aUltrafiltration without chemical aids was tested at these sites and was as effective and less sensitive to changing brine conditions.

Fig. 1. Injection Evaluation Methodology

Fig. 2. Clarification and Processing Methodology

Fig. 3. Evaluation of Headloss Versus Time for Direct/No Chemicals Granular Media Filtration at Constant Flow Rate

Fig. 4. Change in Particle Size Distribution by Multi-Media Filtration with Chemical Pretreatment

Fig. 5. Comparisons of Effluent Quality with Time for the Various Proposed Filtration Schemes

Fig. 6. Comparison of Improved Brine Injectability with Ultrafiltration and Multi-Media Filtration, at Constant ΔP

Fig. 7. Design Schematic for Proposed Downflow Granular Media Brine Treatment System