

CHEMICAL REACTION BETWEEN RECHARGE WATER AND AQUIFER WATER¹

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ABSTRACT

Chemical reaction between recharge water and aquifer water may influence water quality and aquifer permeability, and may cause fouling in discharge wells. However, evidence suggests that the danger of permeability reduction may not be very great.

Precipitates of calcium and magnesium carbonate and iron compounds are the most likely reaction products.

The amount and distribution of reactive chemicals remaining in solution can be related to the dispersive characteristics of a porous medium. Reactions of various rates are considered.

It may be desirable to prevent reaction. It is suggested that, in some cases, this can be accomplished by emplacing a buffer zone of nonreactive water between the recharge water and the aquifer water.

RÉSUMÉ

La réaction chimique entre l'eau rechargée et l'eau d'une couche aquifère

La réaction chimique entre l'eau rechargée et l'eau d'une couche aquifère peut influencer la perméabilité de la couche et la qualité de l'eau.

Nos études indiquent que la quantité et la répartition de la réaction chimique ont rapport aux caractéristiques de dispersion d'un milieu poreux. Les réactions des divers degrés de vitesse ont été estimées théoriquement, y compris celles qui sont rapides en comparaison des vitesses de coulant et celles qui sont lentes en comparaison des vitesses de coulant.

L'effet sur la perméabilité de plusieurs espèces des précipités qui peut former pendant la recharge a été étudié expérimentalement dans les colonnes du sable.

La théorie et les expériences de laboratoire indiquent que, pour l'accumulation d'eau, on peut éviter les réactions chimiques par l'injection d'une zone d'eau non-réactive entre l'eau d'une couche aquifère et l'eau de recharge qui est réactive chimiquement.

INTRODUCTION

A technical problem encountered during artificial recharge through wells is clogging of the pores of the ground-water aquifer and consequent reduction in intake capacity of the recharge well. Chemical reaction between recharge water and aquifer water has been cited as a possible cause of aquifer clogging when recharging through wells (Sniegocki, 1963; American Water Works Association, 1965), and such reaction would also cause change in water quality. A third possible effect of chemical reaction between recharge water and aquifer water is the deposition of reaction products in the well system and distribution facilities of pumping wells located near the recharge point.

This paper discusses some aspects of mixing and chemical reaction between recharge and aquifer water and a possible means of preventing chemical reaction

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between recharge water and aquifer water under some circumstances. The discussions of some of the subjects included herein are necessarily abbreviated, because of space limitations. The interested reader is referred to the references for more extensive treatment.

MIXING

Recharge water and aquifer water must mix before chemical reaction can proceed.

The mechanical mixing that occurs between displacing and displaced fluids in a porous medium is termed hydrodynamic dispersion.

A satisfactory approximate solution to the differential equation for one-dimensional longitudinal dispersion under the physical circumstances that prevail for most problems of interest and for the conditions

$$\begin{aligned}C(x, 0) &= 0; x > 0 \\C(0, t) &= C_0; t \geq 0 \\C(\infty, t) &= 0; t \geq 0\end{aligned}\quad (1)$$

has been given by Ogata and Banks (1961) as

$$\frac{C}{C_0} = \frac{1}{2} \operatorname{erfc} \left(\frac{x-ut}{2\sqrt{Dt}} \right) \quad (2)$$

in which C = the tracer concentration, t = time, D = the coefficient of dispersion with the dimensions Length²/Time, u = average interstitial velocity of fluid flow, and x = distance along the coordinate parallel to the direction of fluid flow.

CHEMICAL REACTION

The reactions between injected and interstitial water that can cause precipitates to form during deep-well injection of liquid industrial wastes were listed by Selm and Hulse (1960) as:

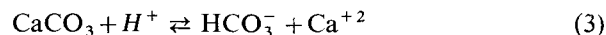
1. Precipitation of alkaline-earth metals such as calcium, barium, strontium, and magnesium, as relatively insoluble carbonates, orthophosphates, sulfates, fluorides, and hydroxides;
2. Precipitation of metals such as iron, aluminum, manganese, chromium, zinc, and cadmium, as insoluble carbonates, bicarbonates, hydroxides, orthophosphates, and sulfides; and
3. Precipitation of oxidation-reduction reaction products.

These reactions are feasible during deep-well industrial waste disposal because of high concentrations of dissolved chemicals in some industrial wastes and in the waters from deeply buried aquifers. However, only a few of the reactions listed above would normally be anticipated during ground-water recharge because ground waters or recharge waters contain only relatively low concentrations of most of the chemicals mentioned except under unusual circumstances. Such is the case where recharge wells are used to establish a hydraulic barrier to sea-water intrusion.

Calcium and magnesium are the alkaline-earth metals frequently found in abundance in ground waters and surface waters. Calcium is the more abundant, except in sea water where the magnesium content is more than 1,000 ppm and the calcium magnesium ratio is about 1:5. Waters from limestones may contain more than 160 ppm

of calcium, and as much as 50 ppm of magnesium is common in waters from magnesian or dolomitic aquifers (Hem, 1959).

The calcium in solution in ground water is often derived from dissolution of calcium carbonate of the aquifer by water containing H^+ ions (Hem, 1960; 1961). The chemical relation is

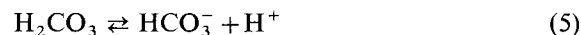


and the equilibrium is

$$\frac{[Ca^{+2}][HCO_3^-]}{[H^+]} = K = 0.97 \times 10^2 \quad (4)$$

in which the bracketed quantities represent activities or, in dilute solutions, concentrations. Unbalancing of the equilibrium shown in equation (4) would lead to precipitation of calcium carbonate. The unbalancing of the equilibrium in a single water is not the problem of concern here, but rather the possibility of obtaining a water supersaturated in calcium carbonate from the mixing of two waters, each one of which is in equilibrium with respect to dissolved calcium carbonate. This problem is examined below.

For the dissociations



and



the equilibrium equations are

$$\frac{[HCO_3^-][H^+]}{[H_2CO_3]} = K_1 = 4.16 \times 10^{-7} \quad (7)$$

and

$$\frac{[CO_3^{-2}][H^+]}{[HCO_3^-]} = K_2 = 4.84 \times 10^{-11} \quad (8)$$

Where the pH is less than about 8.3 the ratio $[CO_3^{-2}]:[HCO_3^-]$ is less than 0.01 and, consequently, equation (7) is sufficient for calculating the amount of dissolved undissociated carbon dioxide. In this case, equations (4) and (7) can be combined to yield.

$$[Ca^{+2}] = KK_1 \frac{[H_2CO_3]}{[HCO_3^-]^2} \quad (9)$$

Using the symbols defined in table 1, equation (9) becomes

$$\bar{X} = KK_1 \frac{\bar{Y}}{(\bar{Z})^2} \quad (10)$$

If it is assumed that the activities or concentrations of H_2CO_3 , HCO_3^- , and Ca^{+2} in solution in the two individual waters (a ground water and a recharge water) can be algebraically combined to yield the activities or concentrations of the ions in a mixture

of the two waters, equation (10) can be rewritten as

$$\bar{X} = KK_1 \frac{\bar{Y} + LY}{\left(\frac{\bar{Z} + LZ}{1+L}\right)^2} \quad (11)$$

in which the amount of recharge water is taken as one unit and the amount of ground water as L units. The assumption mentioned is believed reasonable in the pH range from pH 6 to pH 8, which includes most waters of interest.

Equation (11) expresses the concentration of ions that can theoretically exist in solution under equilibrium conditions. The actual Ca^{+2} concentration immediately after mixing is

$$\bar{X}_{\text{actual}} = \frac{\bar{X} + LX}{1+L} \quad (12)$$

TABLE 1

Symbols used to represent activities or concentrations of chemical constituents in ground water, recharge water, and mixtures of the two waters

Dissolved constituent	Ground water	Recharge water	Mixture of recharge and ground water	Ratio of dissolved constituents
$[Ca^{+2}]$	X	\bar{X}	\bar{X}	
$[H_2CO_3]$	Y	\bar{Y}	\bar{Y}	$Y/\bar{Y} = \beta$
$[HCO_3^-]$	Z	\bar{Z}	\bar{Z}	$Z/\bar{Z} = \alpha$
$[H^+]$	H	\bar{H}	\bar{H}	$H/\bar{H} = \gamma$

If the quantity calculated from equation (11) is greater than that calculated from equation (12), the mixture is considered stable with respect to Ca^{+2} in solution. If the converse is true instability exists and calcium carbonate must precipitate to achieve stability. Thus, for stability

$$KK_1 \frac{\bar{Y} + LY}{\left(\frac{\bar{Z} + LZ}{1+L}\right)^2} > \frac{\bar{X} + LX}{1+L} \quad (13)$$

By using ratios this inequality can be rearranged to yield

$$\beta \geq \alpha \left[\frac{(2+L) + \alpha L}{(2L+1) + \frac{1}{\alpha}} \right], \alpha \geq 1 \quad (14)$$

in which stability exists if the conditions of the inequality are met. Because $\alpha = \beta/\gamma$, it is possible to rewrite equation (14) as

$$\beta \leq \gamma \left\{ \sqrt{\left[\frac{(2L+1)\gamma - (2+L)}{2L} \right]^2 + \frac{\gamma}{L}} + \left[\frac{(2L+1)\gamma - (2+L)}{2L} \right] \right\}, \beta \geq \gamma \quad (15)$$

Plots of equation (15) are given in figure 1 for $L = 0.001, 1, \text{ and } 1000$. In figure 1, for $\beta > \gamma$ and for a given $\beta = \alpha$, $a\gamma < b$ indicates instability for all $L > 0.001$ and $a\gamma > c$ indicates stability for all $L < 1000$. In the region $b < \gamma < c$, the mixture may be stable or unstable depending on L . Mixtures of more than one-half ground water ($L > 1$) are unstable for any $\gamma < d$. Only values of $\beta > 1$ are shown in figure 1, since most recharge waters will contain less dissolved carbon dioxide than will the ground water. Values of $\gamma < 1$ are not shown in figure 1, since for $\beta > 1$ and $\beta > \gamma$ any $\gamma < 1$ indicates instability, and for $\beta < \gamma$ any $\gamma < 1$ indicates stability.

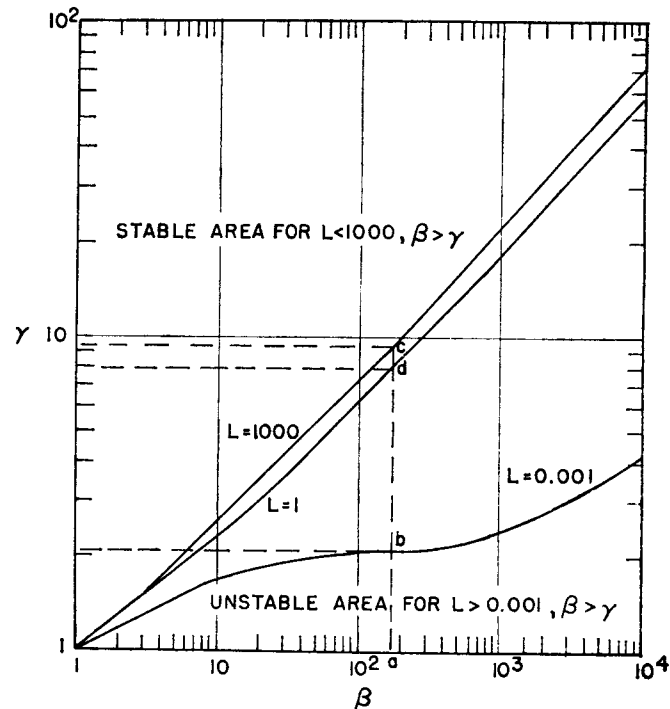


Fig. 1 — Diagram showing stability of a mixture of two waters with respect to dissolved calcium carbonate.

The foregoing analysis shows that when a recharge water stable with respect to dissolved calcium carbonate is mixed with a ground water that is also stable an unstable mixture may result. Furthermore, mixtures may be stable in some proportions and unstable in others. Although the analysis is based on several simplifying assumptions, it is useful for demonstrating general principles.

Dissolved ferrous iron is common in ground waters where concentrations of up to 10 ppm are normal. Rarely, concentrations over 50 ppm may occur in waters with a pH of 5 to 8 (Hem, 1959). Manganese and aluminum are also frequently found in ground waters but usually in very low concentrations.

The principal precipitation reactions associated with deposition of iron in ground water as listed by Hem (1960a) are: (a) precipitation of hydroxides, (b) precipitation of carbonates, (c) precipitation of sulfides, and (d) precipitation of oxidation-reduction reaction products.

Iron in the aquifer water may be oxidized from the ferrous to the ferric state when oxygen or chlorine bearing recharge water mixes with the native ground water resulting in precipitation of the iron as the oxide or hydroxide.

Stumm and Lee (1961) found in laboratory experiments that the rate of oxidation of ferrous iron was first order with respect to Fe^{+2} , was dependent on the partial pressure of oxygen to the first power, and was second order with respect to hydroxyl ion concentration, the overall reaction being fourth order. For purposes of this paper, the oxidation of ferrous iron will be assumed to be dependent on the ferrous iron concentration only, the rate equation being

$$-\frac{d[Fe^{+2}]}{dt} = K[Fe^{+2}] \quad (16)$$

MIXING AND REACTION

Mathematical models for the chemical interaction between injected and interstitial waters can be formulated if it is assumed that mixing is a result of hydrodynamic dispersion and that reaction does not alter the dispersion process.

For reaction that is essentially instantaneous in comparison to the rate of growth of the zone of dispersion, reaction will occur at a vertical interface between the two liquids and no portion of either reactant will ever pass beyond the interface. In this case the distribution of reactants in solution is given by equation (2) up to the interface, where the concentration goes abruptly to zero as shown in figure 2. A purely ionic reaction, such as the reaction of dissolved iron with hydroxyl ions to form iron hydroxide, is essentially instantaneous, although the agglomeration of iron hydroxide molecules into visible aggregates may be slow.

Oxidation of iron from the ferrous to the ferric state is, however, a time dependent reaction and, if the reaction rate is slow in comparison to rate of growth of the zone of dispersion, the reactants will pass beyond the interface before reaction is complete. In this case, mixing must begin before reaction, but thereafter mixing and reaction will occur simultaneously. Figures 3 and 4 show several possible distributions of a chemical in solution at 100 and 1,000 meters away from the bore of an injection well. Curve *a* is the unmodified distribution of a nonreactive tracer. Curve *b* is the distribution of a chemical, for example oxygen consumed during oxidation of ferrous iron, according to a first-order reaction equation. Curve *c* is the approximate distribution that would exist if movement of the injection front or growth of the dispersion zone were stopped until all possible reaction was completed. Curve *b* was obtained by numerical methods too lengthy to describe here. Curve *c* follows the equation (Warner, 1966).

$$\frac{C}{C_0} = \text{erf} \left(\frac{x-ut}{2\sqrt{Dt}} \right) \quad (17)$$

Equation (17) and other similar equations were stated by Warner (1966) to describe essentially instantaneous reaction. Equation (17) in fact describes exactly the case where dispersion proceeds initially with no reaction at all and then, at some point, dispersion

ceases and chemical reaction is completed. This process could be approximated in radial flow, since the flow rate and growth of dispersion zone are both rapid near the bore of an injection well and very slow at a distance from the well bore.

Curves *b* and *c* show that, even at relatively great distances from an injection well bore, time dependent reactions in the zone of dispersion may not be complete and unstable waters could, therefore, be pumped to the surface.

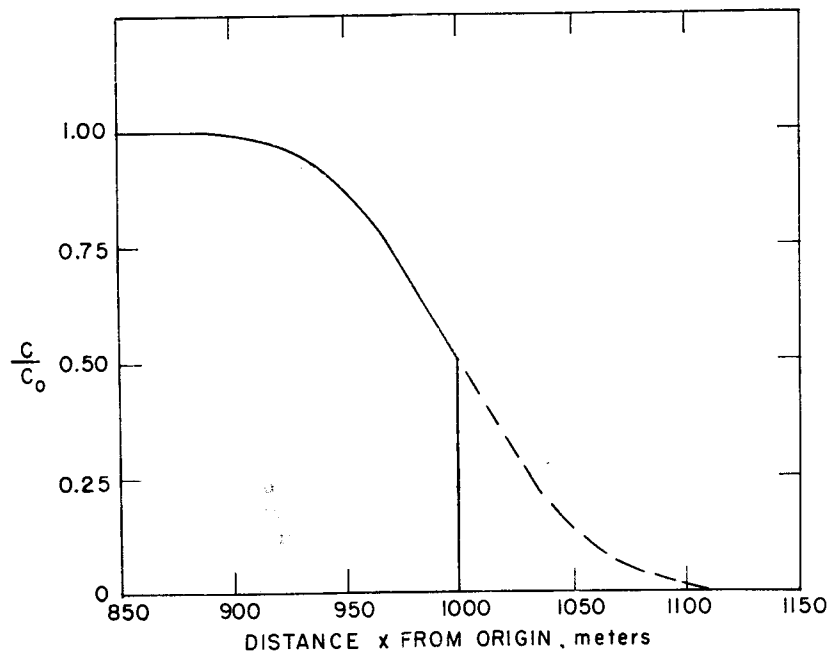


Fig. 2 — Distribution of a reactive tracer in solution 1,000 meters from the origin. Reaction is instantaneous and the reactive chemicals are present in equal normalities. Dashed curve indicates tracer distribution in the absence of reaction. $D' = 1$ meter $u = 1$ meter/day.

This section is presented to show that it is possible to develop a mathematical model to describe the process of simultaneous dispersion and chemical reaction, including time dependency. The major problem thus far has been formulation of a realistic physical model. A detailed explanation of the assumed physical model and mathematical methods will be presented at a later date.

EFFECTS OF REACTION

Laboratory studies by Bernard (1955; 1957) and Warner (1966) showed that, except in one case, little permeability reduction resulted from the displacement of relatively concentrated barium or calcium chloride solutions by sulfate solutions from sandstone cores or sand-packed lucite columns. These findings are attributed to the limited amount of mixing and reaction and to the finely crystalline nature of the precipitates.

Gelatinous iron, manganese, and aluminum oxide or hydroxide precipitates would be anticipated to be more effective in causing permeability reduction than finely crystalline precipitates. Warner (1966) found that the ferric hydroxide precipitate formed during displacement of water containing 26,000 mg/l iron by water containing sodium hydroxide in an equal normality reduced by 30% the permeability of a sand-packed column. Bernard (1957), however, observed no permeability reduction during the displacement of water containing 1,000 mg/l iron by an ammonium hydroxide solution from Berea Sandstone cores.

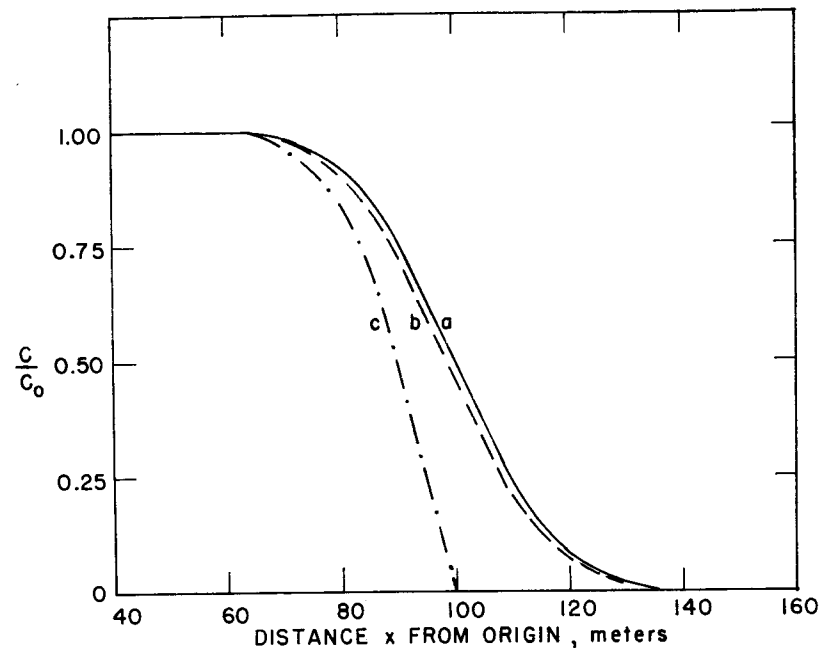


Fig. 3 — Distribution of tracer in solution 100 meters from the origin. Curve *a* is with no reaction. Curve *b* is with first order reaction. Curve *c* follows equation (17). Reactive chemicals are present in equal normalities. $D' = 1$ meter, $u = 1$ meter-day, first-order reaction constant $K = 10^{-6} \text{ min}^{-1}$.

The above cited laboratory studies indicate that the danger of permeability reduction resulting from mixing of incompatible waters during ground-water recharge may not be as great as has been suggested by some writers. This conclusion is substantiated by the lack of evidence of aquifer plugging, during oilfield waterflooding operations (Bernard, 1957) or during ground-water recharge operations (Steinbruegge and others, 1954).

In addition to the mixing during dispersion within an aquifer, recharge water and native ground water may mix in the well bores of discharging wells that are near the source of recharge. It seems possible, based on the earlier discussion of the stability of water mixtures, that unstable mixtures could be formed at a discharge well, resulting in deposition of calcium carbonate or iron compounds in pumping or

distribution systems. This could also occur as a result of mixing within the aquifer if reactions were not complete at the time the mixed water was pumped to the surface. Gates and Caraway (1965) discussed the formation of barium sulfate precipitates in producing oil wells in the Wilmington oilfield, California. The unstable water in this case resulted from mixing sulfate bearing waterflood injection waters with aquifer water that contains barium.

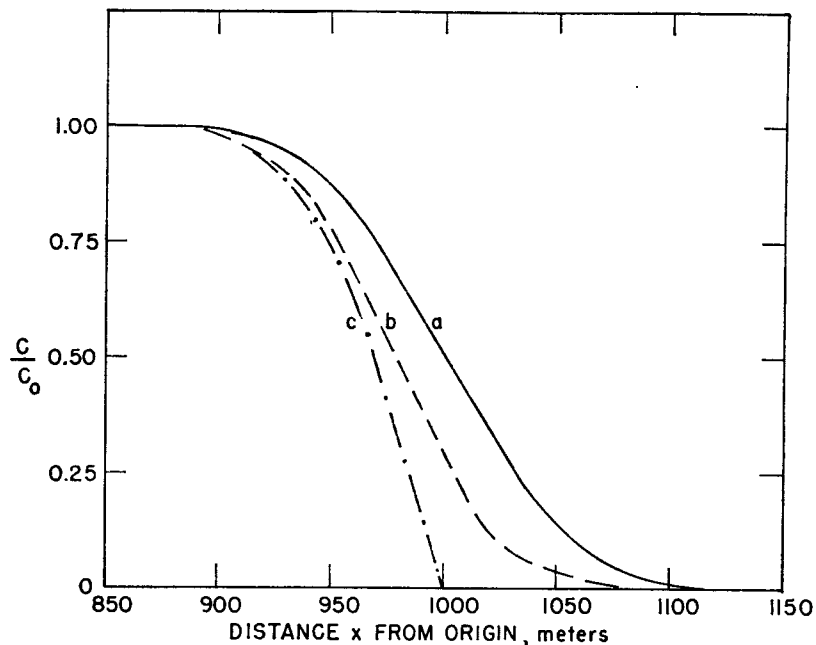


Fig. 4 — Distribution of tracer in solution 1,000 meters from the origin. Curve *a* is with no reaction. Curve *b* is with first order reaction. Curve *c* follows equation (17). Reactive chemicals are present in equal normalities. $D' = 1$ meter, $u = 1$ meter/day, first-order reaction constant $K = 10^{-6} \text{ min}^{-1}$.

PREVENTING OR REDUCING REACTION

Preinjection treatment to make recharge water nonreactive is a means of preventing chemical reaction between recharge water and aquifer water. Another possible means of preventing reaction is the injection of a zone of nonreactive water between the aquifer water and the main body of injected liquid.

It is believed that a buffer zone may be useful in preventing reaction out to a point where any clogging that does occur will have little influence on the intake rate of the injection well. For example, a buffer zone could be used to prevent reaction out to the point of stagnation upgradient from a recharging well.

If water is injected at a constant rate Q into an aquifer of thickness h with a natural hydraulic gradient I and permeability K , the distance x that the water will eventually

travel upgradient is given by the equation (Todd, 1959, p. 86):

$$x = \frac{Q}{2\pi KIh} \quad (18)$$

Warner (1966) showed that, in purely radial flow, the quantity of water V_b that will form a buffer zone to prevent reaction to a distance r from an injection well can be calculated from the equation:

$$V_b = 217.1 \sqrt{D'r^3} h\theta \quad (19)$$

in which the coefficient of dispersion $D' = D/u$ with units of Length, $\theta =$ the porosity, the units of length are in feet, and the volume is in gallons.

For the conditions

$$\begin{aligned} Q &= 300 \text{ gpm (1.14 m}^3\text{/min)} \\ K &= 1,000 \text{ gal/day-ft.}^2 \text{ (40.7 m}^3\text{/day-m}^2\text{)} \\ I &= 10^{-3} \\ h &= 100 \text{ ft. (30.5 m)} \end{aligned}$$

x in equation (18) = 688 ft. (210 m).

For $D' = 1$ ft. (0.305 m) and $\theta = 0.3$, V_b in equation (19) = 117×10^6 gal ($0.44 \times 10^6 \text{ m}^3$). About 270 days of continuous injection at 300 gpm would be needed to emplace the required buffer zone.

CONCLUSIONS

Chemical reactions between recharge water and aquifer water may alter the chemical quality of the water in the zone of reaction, cause fouling of pumping and distribution facilities in pumping wells near the recharge point, and perhaps lead to permeability reduction in the ground-water aquifer. Existing laboratory and field data suggest, however, that the danger of permeability reduction may not be very great.

Precipitates of calcium and perhaps magnesium carbonate and various iron compounds are thought to be the most likely reaction products. Theoretical analysis suggests that calcium carbonate could be precipitated from mixtures of two waters both initially stable with respect to dissolved calcium carbonate. Dissolved ferrous iron in native ground water could be precipitated as ferric oxide or hydroxide as a result of mixing with oxygen-bearing recharge water. This reaction is a time-dependent one.

Mathematical models that describe the amount of chemical remaining in solution when chemical reaction occurs between recharge and aquifer water can be formulated from the basic dispersion equations if it is assumed that reaction does not alter the dispersion process. The models that have been obtained show the distribution of chemicals in some selected cases.

Reaction between recharge water and aquifer water can be prevented by continuous pretreatment to make recharge water nonreactive. A possible alternative method of preventing reaction in some cases is injecting a buffer-zone of nonreactive water between recharge water and aquifer water.

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ECONOMICS OF ARTIFICIAL RECHARGE FOR MUNICIPAL WATER SUPPLY

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ABSTRACT

Ever-growing demands for fresh water have forced a re-examination of the methods for controlling water quality. Reclamation of waste-waters, as an alternative to conventional methods of developing water supplies, is gaining more importance as a means of better utilizing the considerable investment in waste treatment facilities, usually required by law merely to permit effluents to be discharged. Further marginal investment resulting in reclaiming wastes for economic use represents a sound means for achieving both a qualitative and quantitative conservation of resources and eliminates the detrimental effects of discharging partially treated effluent to receiving waters.

A research project was undertaken to determine whether or not waste reclamation could be economically competitive with other water sources for municipal water supply. Numerous advanced waste treatment systems and recycle schemes were evaluated. Waste-water renovation through ground water recharge proved to be the most feasible solution to reclamation of the effluent of any type treatment plant today.

Further study has evaluated the chemical and physical limitations of artificial recharge using municipal wastes; the economic trade-offs between additional treatment prior to recharge and greater land utilization; and the break-even point for land values as a function of economies of scale. Finally the economics of a particular case-study in the arid West of the United States is discussed as well as a proposed scheme for converting the nation's capital, Washington, D.C., in the humid-East from using solely surface water supplies to using artificial recharge of undeveloped aquifers for future expansion of water supplies.

RÉSUMÉ

Les besoins sans cesse croissants en eau fraîche ont nécessité une reconsidération des méthodes de contrôle de la qualité de l'eau. L'utilisation des eaux usées comme une alternative en compétition avec les méthodes habituelles de développement des utilisations d'eaux, a gagné de l'importance comme un moyen de mieux utiliser les investissements considérables faits pour l'épuration des eaux usées en vertu des exigences légales pour permettre le déversement de ces eaux dans les cours d'eau. Les investissements marginaux requis pour la réutilisation de ces eaux conduisent à une conservation quantitative et qualitative de ressources et éliminent les effets nocifs d'une restitution d'eaux partiellement traitées aux cours d'eau.

Une recherche a été entreprise pour déterminer si la réutilisation des eaux usées pouvait être économiquement compétitive vis-à-vis d'autres sources d'eau pour usage municipal. De nombreux systèmes perfectionnés d'eaux usées, et des schémas pour recycler ces eaux ont été évalués. La rénovation des eaux usées par la recharge des nappes s'est montrée la solution la meilleure que tout autre type de traitement.

Des recherches plus poussées ont évalué les limitations physiques et chimiques de la recharge utilisant des eaux usées; l'économie d'un traitement préalable des eaux usées avant la recharge a été examinée.

Enfin, l'économie d'un problème particulier dans l'Ouest aride des Etats-Unis est discutée, comme d'ailleurs un plan de conversion de l'alimentation en eau de la capitale fédérale, Washington, D.C., dans le Nord-Est humide en faisant participer la recharge de nappes peu développées à cette alimentation.

INTRODUCTION

The possibility of increasing the supply of usable water depends no longer upon the exploitation of new sources but upon the interplay of technological change and

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