

GROUNDWATER IMPACT OF SILICATE GEL INJECTIONS

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ABSTRACT

The increasing application of chemical soil consolidation techniques by silicate gel injections always involves the risk of changing the surrounding groundwater/soil environment, when these are introduced into an aquifer and thus submitted to an intense process of solution. These impacts on groundwater related to the preparation of the silicate gel solutions on the one side and their characteristics on the other are:

- 1) An increase of alkalinity due to alkaline components of the water glass or alkaline and alkaline-earth precipitants,
- 2) an increase of the organic content in the case of an application of organic precipitants. This causes O₂ consumption and reduction of the O₂-containing compounds leading to a transient strongly reduced groundwater medium,
- 3) an increase of the heavy metal content due to impurities of the water glass or dissolution from the sediment by the action of CO₂,
- 4) sulphide precipitation of heavy metals in a reduced medium only.

Besides, technical and human failures may furthermore raise the estimated environmental pollution.

INTRODUCTION

This contribution is intended to deal with a particular problem of groundwater and soil hygiene caused by a technique of special underground works which has undergone an increase in scale and importance since its introduction in 1925: the chemical consolidation of foundation soils by injection of silicate gel.

These techniques work on the basis of an injection of water glass solutions and precipitants. In cohesiveless, granular foundation soil formed by sands of fine to intermediate grain, the application of such techniques will result in a compaction with a simultaneous increase in shear-strength and the sealing of interstices (refs. 1,2).

Since their introduction in 1925 until 1966, only about 170 000 m³ of soil have been chemically consolidated with these special foundation techniques (ref.3). Today some 10⁵ m³ soil/a will be consolidated with another 10⁴ t of chemical solutions in the Federal Republic of Germany including Berlin (West).

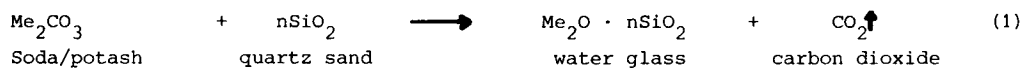
Nevertheless, the related impact on groundwater quality caused by the direct injection of silicate gel solutions into the aquifer has been evaluated insufficiently till now.

GROUNDWATER IMPACT RELATED TO THE PREPARATION AND CHARACTERISTICS OF SILICATE GEL SOLUTIONS

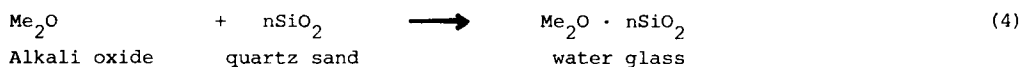
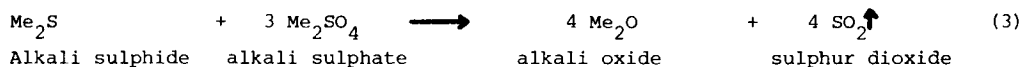
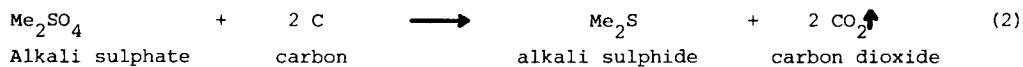
When taking into consideration the chemical and physical characteristics of the basic component water glass and the different reactants, hygienic effects on groundwater due to silicate gel injections become most evident.

Water glass solutions are concentrated silicic acid solutions with a definite, but not stoichiometric, fraction of alkali, often sodium or potassium. The chemical industry distinguishes between soda and potash glasses on the one hand (1) and sulphate glasses on the other (2-4) according to the production process (ref. 4):

Soda/potash glass



Sulphate glass



Soda and potash glasses are directly extracted from a melt of soda or potash and pure quartz sand. This reaction is nearly quantitative as carbon dioxide is expelled by silicic acid (1). The prefix "n" stands for the molar ratio of silicic acid to alkali oxide.

For economic reasons, carbonates being alkali components are sometimes replaced by sulphates. Then the sulphate will at first be reduced to sulphide by carbon (2). Sulphide will be oxidized to sulphur dioxide and alkali oxide by alkali sulphate added in excess (3). Both reactions are irreversible because of one product being expelled as a gas (CO₂ and SO₂). Finally, the extremely reactive alkali oxide will completely react with quartz sand to form water glass (4).

The residual fraction of 1 - 1.5 % sulphate in sulphate glasses is quite considerable if compared with 0.1 - 0.2 % sulphate contamination of soda glass.

From both processes water glass is extracted as solid clear glass pieces ("Stückengläser") of different colour according to their impurities. They have to be solved in water to get fluid water glasses.

Fluid water glasses are extremely alkaline as they are solutions of a basic compound. Nevertheless, industrial producers sometimes distinguish between sodium water glasses as highly silicic, neutral and alkaline ones according to their alkali fraction. Potassium water glass is produced by a few specifications only and distinguished as potash and potassium sulphate glasses (ref.5) (Table 1).

Table 1

Varieties of water glass (calculated for "Stückenglas") (ref. 5)

	Me ₂ O % w/w	SiO ₂ % w/w	SiO ₂ :Me ₂ O n	Me ₂ SO ₄ % w/w
highly silicic glass	20	80	3.8 - 4.1	0.1
neutral soda glass	23	76	3.4 - 3.5	0.1
neutral sulphate glass	22	73	3.4 - 3.5	1.5
alkaline soda glass	32	66	2.1 - 2.2	0.1
potash glass	28	71	3.9 - 4.1	0.2
potassium sulphate glass	25	64	3.9 - 4.2	1.0

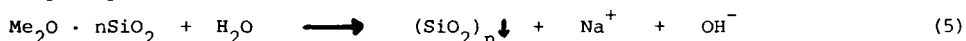
The pH values of all water glass solutions vary between 11 and 13 (ref. 6). Even in extensive dilution as the injection into an aquifer would imply, the pH value of the vicinity decreases slowly according to its buffer-like characteristics. The following example of a so-called "neutral" soda glass makes this quite obvious (Table 2).

TABLE 2

pH changes at different dilution steps of a sodium water glass solution (neutral soda glass) (ref. 4)

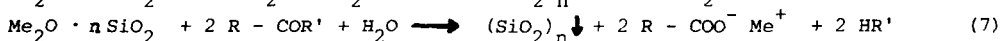
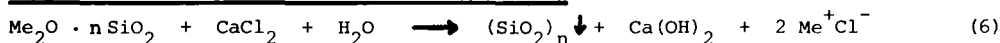
Fluid glass; n = 3.40	g/l	100	10	5	1	0.1
Sodium silicate	g/l	35.8	3.58	1.79	0.36	0.04
pH value (20° C)	$-\log \frac{[H^+]}{[OH^-]}$	11.4	11.0	10.8	10.8	9.7

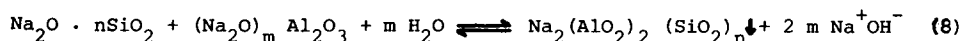
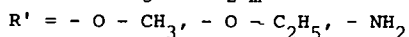
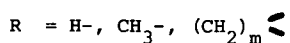
Release of soda or potash lye by precipitation of an insoluble silicic acid gel is a hydrolysis reaction (5) (ref. 7):



A complete reaction theoretically releases 150 - 500 kg soda lye/t sodium water glass according to the solid and alkali fractions. Extensive building projects usually imply injection of some thousand tons of water glass solutions.

Pure water glass solutions are converted from the sol to the gel state by appropriate specific reactants, e.g. alkaline earth ions (6) (ref. 7), organic compounds (7) (ref. 8) or sodium aluminate (8).





Neutralization by adding cations is the main reaction (6) of the JOOSTEN technique which was the first pure silicate gel technique developed in 1925. Concentrated water glass solutions are precipitated by calcium or magnesium chloride solutions producing extremely hard gels. Beneath the silicate gel a slightly soluble alkaline calcium or magnesium coating forms in the soil interstices. So application of this technique will affect groundwater quality by an increase of the electrolyte content as well as of the pH value of the groundwater.

Nevertheless, using divalent cations will mean a technical disadvantage as the abrupt gel formation excludes simultaneous working with two injection solutions.

Modern techniques of hard gel injections are based on the extraction of hydroxyl groups (7). They use carbonic acid derivatives, whose hydrolysis is a time-dependent reaction consuming hydroxyl ions. The available time of relaxation can be adjusted according to the time necessary for the simultaneous injection of water glass and hardener after mixing both reactants.

Common reactants among carbon acid derivatives are especially ethyl acetate, technical more or less pure mixings of dicarbonic acid methyl esters and formamide. The reaction principle of all substances is the same but they are distinguishable in their economy and application as well as in their degree of technical purity and characteristics of the hydrolysate.

In addition to the soda lye that is released, a remarkable quantity of organic substance is introduced into the groundwater. Most of this organic matter will be microbially decomposed leading to a depletion and consumption, respectively, of the natural oxygen content of the groundwater. At last oxygen-containing compounds such as sulphates and nitrates are reduced. The following scheme shows the possible effects of a water glass/ethyl acetate injection into an aquifer:

Soda lye	→	pH shift to alkaline range
Organic substance	→	high O ₂ demand for its decomposition
Decomposition process, O ₂ -demand	→	CO ₂ formation, decreasing oxidation-reduction potential (ORP)
CO ₂ -formation	→	pH shift to acid range
Decreasing ORP	→	reduction processes: NO ₃ → NO ₂ → NH ₄ ⁺ Mn and Fe oxides → Mn ²⁺ and Fe ²⁺ ions, SO ₄ → S → H ₂ S
Sulphide sulphur formation	→	precipitation of heavy metals as sulphides, pH shift to neutral or even alkaline range

Precipitation of heavy metals as sulphides which had been introduced by impurities or which became dissolved from the sediment by the action of CO₂ is reversible in the

case of a normalization of the groundwater medium. Hence temporarily increased heavy metal contents may dissolve somewhat later in connexion with an impurity of the reactants or heavy metal containing sediments (e.g. marine deposits).

According to the "List of Hazardous Substances in Water" (ref.9), the reaction products of the carbonic acid derivatives, especially the reaction products, ammonia, methanol, formic and acetic acid or formate and acetate as well as soda lye, have to be assigned to water hazard class 1 which identifies them as slightly hazardous for water.

Quantities of organic compounds used in injection techniques depend on the water glass specification and the degree of neutralization. So e.g. 100 - 900 kg ethyl acetate/t water glass are necessary for a neutralization of 40 - 80 %.

When using formamide, the groundwater and the environment of the construction site are exposed to ammonia as mentioned before. As formamide itself is teratogenic, this chemical is a problematic one for injection techniques (ref.10).

Another common technique is the simultaneous injection of highly diluted water glass solutions and sodium aluminate (8). The silica gel again flocculates in the soil interstices after an adjustable time of relaxation. The risk of groundwater contamination by using this hardener could also be seen in the increase of alkalinity of the groundwater in the vicinity of the injection while other by-products and technical impurities are of minor rank.

Water glass sodium aluminate injections are only used for the production of hydrous soft gel provided for sealing techniques. Therefore, sodium aluminate is a technical completion but not alternative to other reactants.

GROUNDWATER IMPACT CAUSED BY TECHNICAL AND HUMAN FAILURE

The environmental consequences of technical processes do not only depend on the estimated adverse effects but to a certain extent also on the possibility of technical breakdowns or human failure.

There have been incidents in the Federal Republic of Germany and Berlin (West) which made the possibility of an extraordinary contamination of water, soil and air in the vicinity of injection sites obvious.

In 1974, injection material emerged uncontrolled in the cellar of a construction site and was pumped by the contractors into a near-by depression. So 5 m³ of soil were permeated and contaminated. In spite of the risk of groundwater pollution due to seepage, the contaminated soil was not moved to a suitable waste disposal site until 4 months later.

In 1978, infiltration of about 2.5 - 3 m³ strongly alkaline groundwater into a lift shaft became known from another place. A three-year-old injection was found to have been responsible for the contamination of groundwater and ambient air as it had been constructed in a cone of influence not taking into account the rise of the

water-table in 1978 above the level of the injection. Leaching of ammoniacal substances from the injection therefore took place with a time lag.

At another place, in 1978 too, a big quantity of seepage extremely contaminated by injection material was pumped out of an excavation directly into a storm-water discharge emptying into a receiving water.

However, as a remarkable consequence, permission for chemical injections into soils in the area studied has been made obligatory for the first time. A similar development is noted for other administrative areas. Also contractors for underground works and scientific institutions are looking for appropriate and more favourable methods to eliminate avoidable groundwater contamination in the future.

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