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Thermodynamic modeling of silica dissolution kinetics of quartzitic aggregates stored in highly alkaline solution

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Abstract

Knowledge's on the dissolution behaviour of SiO_2-containing aggregates are the basis for understanding damaging processes regarding ASR. Because of many influences, the solubility of aggregates has still needs to be determined by dissolution experiments. This article is about the reliably modeling of silica dissolution of relatively pure SiO_2 containing aggregates in alkaline solutions. Therefore, quartz, quartzite, opal sandstone and flint with different silica modifications were chosen. A kinetic model based on the surface area was derived and numerous thermodynamic data from different authors were evaluated.

The surface area of the aggregate grains were determined by an adapted BET method. For this measurement, the original grain sizes were used as they are also used so in concrete. These aggregates were stored in 0.1 mol and 1.0 mol KOH solution at 40°C and 80°C respectively. The dissolution experiments too were performed with grains in the original particle sizes. The concentration of silica in the liquid phase was measured by ICP-OES.

The model based on a comparison of calculated and experimental determined silica concentrations, because some parameters of the differential equations are still unknown. The model includes the temperature and molarity dependence of the silica dissolution. The model also takes into account the influences of the SiO_2 structures and can vary between crystalline and amorphous SiO_2 in a wide range. Thermodynamic data suitable for modeling are recommended.

In future, these dissolution calculations shall be extended to other non-pure quartzitic aggregates. Instead of time-consuming ASR concrete prism tests the silica dissolution rates may be used then to indicate quickly the ASR sensitivity of aggregates. Of course, it will be possible to distinguish between innocuous, medium and highly reactive grains.

Keywords*: alkali silica reaction; kinetic modeling; quartzite aggregates; silica dissolution; thermodynamic data*

1. INTRODUCTION

The authors of this paper have dealt with solubility experiments of aggregates for many years. A large amount of solubility values of aggregates in highly alkaline solutions exists which demands a deeper evaluation regarding the description of the dissolution kinetics of silica. Especially, the extensive existing data sets should make it possible to calculate the dissolution behaviour of aggregates, which consist mainly of SiO_2.

Already in the 80s of the past century, dissolution experiments were done especially with quartz at different temperatures and pH values. Knauss and Wolery [1] reported on the determination of the dissolution kinetics of quartz as a function of pH and time at 70°C. Other researcher [2] investigated the influence of organic acids to the dissolution and found that the solubility of quartz is strongly increased. However, the results indicated that the concentrations were maintained far from equilibrium with respect to quartz. For the calculation, they used the geochemical modeling code EQ3/6, developed by Wolery. However, his work based on former investigations from the 60s and 70s of the past century. The knowledge of silicate dissolution mechanisms were of great importance especially for geochemical processes which meant that mostly the solubilities were determined only in weakly alkaline environment until pH of 11. Dove [3] described the dissolution kinetics of quartz in mixtures of different salts in near-neutral pH solutions over a wide temperature range. Already since 1884 the nature of silica dissolution to terrestrial weathering was of great interest. More than 130 years of research delivered many results of dissolution values, however a practical use of understanding what happens with quartz in a very highly alkaline environment, is still missing. In [4] dissolution experiments of quartz are described where the concentration of 0.02mol/kg solution do not exceed. With such experiments the formation of three monomeric silica species H_4SiO_4 (for neutral conditions), $H_3SiO_4^-$ and $H_2SiO_4^{2-}$ (for alkaline conditions) have been dedected. This is still the basis for most kinetic calculations today, also here in this paper. The reason for this is simple because thermodynamic data exist for these species from many researchers. A selection is part of the reference list [16-21].

In the 90s Rimstidt [5] performed new quartz solubility tests to compare it with information from the literature. Revised thermodynamic properties of H_4SiO_4 specie were published to calculate the solubility of amorphous silica with 116 ppm and with 6 ppm for quartz at 25°C.

Bickmore [6] went another way to describe the dissolution of quartz. The basis here were experimental data of quartz dissolution experiments in a wide range of temperature (25-300°C) and of pH (from 1-12). The model rate law, a mechanistic rate law, was an e-function, which included the temperature and best fitted reaction enthalpy values for different silanol groups. He summarized his work with the statement that further investigations require the development of more predictive and realistic models of surface speciation. Exactly this is what will be done in this paper.

Jehn [7] showed in her work that especially very dense aggregates dedicate a surface controlled reaction mechanism. Certain highly silica-containing materials (rock crystal, opal sandstones and flints) with different densities were investigated. The reaction of the high dense materials (HD) could be understood in a good way without any consideration of transport processes inside of grains. That`s why the knowledge of the specific surfaces of natural grains have a great importance for understanding what happens with aggregates in highly alkaline solutions. Different methods exist, however it seems that the BET method provides more satisfactory results in comparison for example to the CT-method [8].

Another difficulty is that natural quartz grains consist not only of well-crystallized quartz but also of strained quartz zones. A typical example is quartzite, a metamorphic rock with deformed crystallite zones and edges. Additionally, a further problem is that many rock materials, which are used as aggregates in concrete, contains aluminium in detectable soluble amounts. Al can control the silica release [9], but this mechanism shall not be considered here in this paper.

The aim is to develop a relatively simple computer program, based on thermodynamic data and kinetic laws, which makes it possible to calculate the solubility of natural aggregates only from the knowledge of the chemical and mineralogical composition and of surface values. Kinetic models are described in different variations. Merkel et al. [10] mention an equation calculating the reaction rate of minerals. This equation contains a term k_r (reaction rate constant), a term A_o (for the surface of minerals) and a term V (for the volume of the solution). Other equations base in general on the same fundamental parameters and are published and summarized for example by Sigg and Stumm [11]. A computer-based model for calculation of dissolution and precipitation processes in aqueous systems was developed by Parkhurst et al. [12], called PHREEQC. This program implements thermodynamic and kinetic parameters of many species in aqueous solutions, mostly to calculate changes in such systems for a geochemical use. At high pH values, the model reaches its limits. In the last years, Crundwell has published many papers, which dealt with dissolution processes of minerals. In [13] he introduced the theory of the surface potential difference between the solid surface and the liquid phase outer the Stern layer. From this theory, he could derived a model of quartz and silica dissolution which predicts the observed orders of reaction. He summarized the models and equations of Rimstidt and Bickmore mentioned above.

While many researchers investigated relatively pure materials (pure quartz, amorphous silica) already Labrid [14] dealt with the dissolution behaviour of natural sand. He developed an equation system, which was able to describe the solubility of the sand fraction in dependence on the pH and the equilibrium state between dissolution and precipitation of Si-monomers on the surface. The approach of Labrid was here further developed under consideration of changed surfaces of grains during the dissolution process.

2. MATERIALS AND METHODS

2.1 Experimental

For the experimental investigations four different, highly SiO_2 containing aggregate materials were used. As a very well crystallized material, quartz (rock crystal) was chosen. The second material was a quartzite, a metamorphically embossed quartz rock. To consider the influence of amorphous SiO_2 structures on the dissolution behaviour, an opal sandstone and a flint rock were also stored in alkaline solutions. All materials chosen consist of more than 95wt.-% of SiO_2 except the opal sandstone (approx. 80wt.-%), but not or in hardly measurable quantities of alumina only. The composition of aggregates used is summarized in chapter 2.2.2. The dissolution experiments were performed with the original grain sizes 2-8 mm (for rock crystal 4-8 mm) and 8-16 mm respectively. Explanations for this are given in chapter 2.2.1.

The grains were stored in 0.1M KOH solution at 40°C and 80°C and 1.0M KOH solution at 40°C, the last storage condition corresponds to the condition of the CPT in Germany. 90g of each material with the corresponding grain size was put in 900ml KOH solution. Three ml solution was removed from the storage vessel at each analytical time. The times are freely selectable between zero and almost 365 days. The solution volume must decrease by this and this is taken into account in the calculation too. During the storage, the vessels were not shaken. The samples were only briefly moved shortly before the solution was removed. The silica concentrations were determined using an ICP-OES spectrometer from Thermo Fisher Scientific company. The measured SiO_2 concentrations in the solutions are the basis for the calculations.

2.2 Kinetic model

2.2.1 Determination of the specific surface

The model used is a surface controlled model which means that no transport mechanisms inside of grains are considered. Selected were rock materials with a high density and a very low porosity to guarantee that interactions between the highly alkaline solution and the aggregate grains can only occur on the surface area and other areas, which are only accessible from the outside. A very important parameter for this model is the specific surface of the grains used in the dissolution experiments. The specific surfaces were determined using the BET method with nitrogen gas.

Normally this method is applied to investigate powdery samples with a high specific surface. Normally the powdery samples are filled from above through a capillary (6 mm diameter) to the spherical measuring cell at the bottom. In opposite the original grains (2-8 mm, 8-16 mm) do not fit through this capillary and additionally the volume of the spherical cell is too small for the large grains. To realize relatively good measurement conditions, a larger sample vessel with a separate acces was tested. The capillary remained the same but the cell volume was larger and could accommodate several full-size grains. The detectable surface is limited by 0.5 m^2/g. The measurement of the specific surfaces was repeated three times in all cases. The glass capillaries with the volumetric flask (left spherically, right cylindrical) can be seen in Figure 2.1.

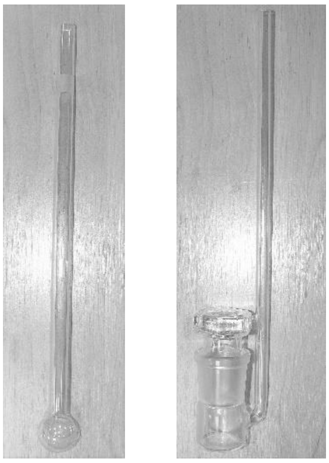

Figure 2.1: Equipment to measure the specific surfaces of original grains

2.2.2 Mineralogical and chemical composition of the aggregate material

In a first step, the investigations were concentrated on relatively pure SiO_2 containing rock materials. Selected were:

- a rock crystal (quartz) with 100% crystallinity and of course 100% SiO_2, the crystallinity index is per definition 10,
- a quartzite rock material with approx. 98-99wt.-% quartz and 1-2wt.-% muscovite (the chemical composition is approx. 99%SiO_2), the crystallinity index for 2-8mm grains is 7.97, for 8-16 mm material 6.89,
- a flint with 97wt.-% SiO_2 and a mineralogical composition: 54wt.-% quartz, 46wt.-% amorphous and cristobalite
- an opal sandstone with 80wt.-% SiO_2 and a composition: 67wt.-% quartz, 14wt.-% amorphous phase and cristobalite, 13wt.-% limestone and a little bit feldspar and hematite (the crystallinity is more or less unknown).

Especially the flint and the sandstone material consist of different SiO_2 modifications, which has to be considered in the model.

2.2.3 The basic kinetic module of the model for SiO_2 dissolution

The model bases on the following Equation system:

$$\left.\begin{aligned} \frac{dc_{Si}}{dt} &= \frac{k_{1,Si} * \mathrm{A}_{\mathrm{Si,eff}}(t)}{\mathrm{V}} * \left(1 - \frac{c_{Si}(t)}{\mathrm{c}_{\mathrm{Si,eq}}(t)}\right) \\ \frac{dA_{Si,eff}}{dt} &= A_{Si}(t) - \frac{c_{Si}(t) * A_{Si,t=0}}{n_{Si}(t)} \\ \frac{dc_{Si,eq}}{dt} &= \frac{dOH^-}{dt} \\ [OH^-] &= [K^+] + [H^+] - 2[H_2SiO_4{}^{-2}] - [H_3SiO_4{}^{-}] \end{aligned}\right\} \tag{1}$$

with:

dc_{Si}/dt = change of the SiO_2 concentration as a function of time [mol/(l*d),

$A_{Si,eff}$ = for dissolution processes effective surface [m^2/g], changes with time t,

V = volume of the alkaline solution (here 900ml), changes with each withdrawal,

$k_{1,Si}$ = velocity constant of the silica release [1/d],

c_{Si} = SiO_2 concentration of the solution at a certain time t [mol/l],

$c_{Si,eq}$ = thermodynamic equilibrium concentration of SiO_2 (depends on the SiO_2 structure and changes with the OH^- concentration),

n_{si} = represents the number of SiO_2 monomers on the mineral surface, expressed in concentrations, contains conversion factors to recalculate surfaces in concentrations

dOH^-/dt = change of the OH^- ion concentration during silica release.

Another important value is the volume of a SiO_2 monomer in the solid phase, which can be found in the literature with: $V_{SiO2} = 4.5*10^{-23}cm^{-3}$ [15]. Based on this value, the starting surface and the changes during silica release expressed as concentrations can be calculated assuming a tetrahedral structure.

The differential Equation system (1) can be solved by numerical integration systematically with the following starting condition: at time t = 0d the SiO_2 concentration c_{Si} is also zero. Each calculation step delivers a dissolved SiO_2 concentration, which can be recalculated from a change of the surface and a change of OH^-. The new surface and the new OH^- concentration enter into the next calculation step. Additionally the equilibrium concentration $c_{Si,eq}$ determined by thermodynamic calculations also changes because it depends on the OH^- concentration too (which means on the pH value). Important is this correlation for materials, which release large amounts of silica into the alkaline solution (opal sandstone, flint). It follows that equilibrium values for SiO_2 can only be specified for the initial conditions. Activity coefficients using Davies equation (for ion strength I until 0.5mol/kg) and the WATEQ-Debye-Hueckel (until I= 1mol/kg) equation (equations can be found in [10] and [11]) are also taken into account in the calculations.

The calculation procedure considers the silica species, which are stable in dependence on the pH value of the alkaline solution. Figure 2.2 demonstrates the distribution of SiO_2 species over a pH range from

6 to 14. From a thermodynamic point of view stable are approx. 80% $H_3SiO_4^-$ and 20% $H_2SiO_4^{2-}$ at pH 13. However, this ratio changes with changes of pH.

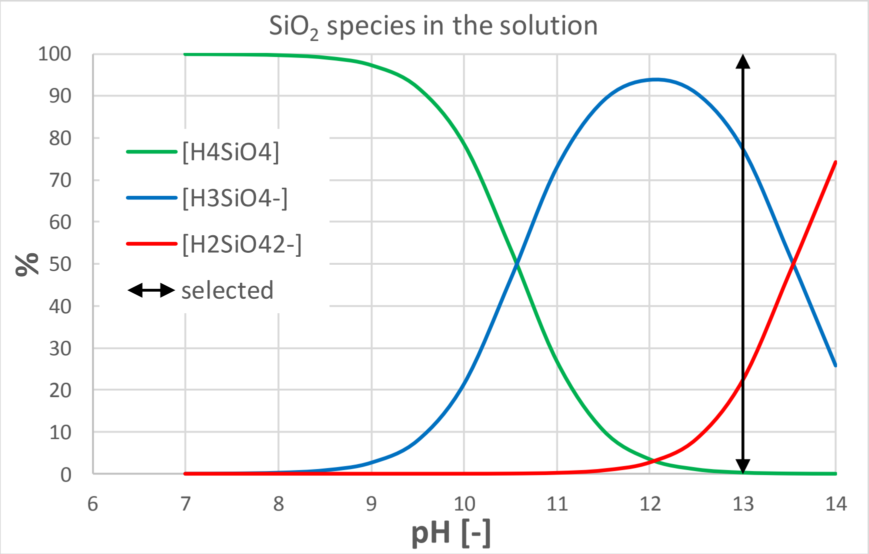

Figure 2.1: SiO_2 species in the aqueous solution as a function of pH, for equations see [16] (the selected pH value is marked)

3. RESULTS

3.1 Calculation for rock crystal

The dissolution experiment of rock crystal was performed until more than 300 days. The shape and the size of the crystals can be seen in Figure 3.1. The next figure represents the results of the measured SiO_2 concentration and the calculated values according to the differential equation system.

Figure 3.2 gives the calculation results for conditions 0.1 M and 80°C, Figure 3.3 for 0.1 M and 40°C.

Figure 3.1: natural rock crystal (picture from [7])

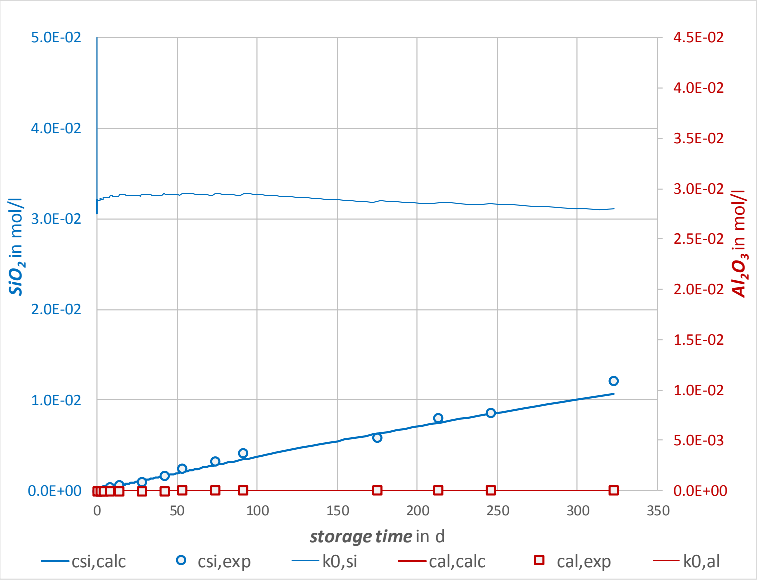

Figure 3.2: Measured data and calculated curve of rock crystal material (grain size 4-8 mm) for 80°C and 0.1 M KOH

The following parameters are valid for this calculation:

$A_{Si}(t=0) = 0.21\ m^2/g$,

$k_{1,Si} = 8.63*10^{-2}\ d^{-1}$,

$c_{Si,eq} = 1.1*10^{-1}$ mol/l (for rock crystal in 0.1 M KOH, 80°C)

The Figure contains also the development of the SiO_2 equilibrium concentration, actually the activities. This value depends on the thermodynamic data, but also on the change of the hydroxide concentration caused by silica release. It can be clearly seen that the soluble silica concentrations are far away from the equilibrium under these conditions.

The same calculation can be made for the system 40°C and 0.1 M KOH solution until more than 300 days too.

In both cases the calculated curves can be fitted be variation of $k_{1,Si}$, the velocity constant of the silica release [1/d]. As expected, this value is higher for conditions 0.1 M KOH and 80°C than for 0.1 M KOH and 40°C because of the higher temperature.

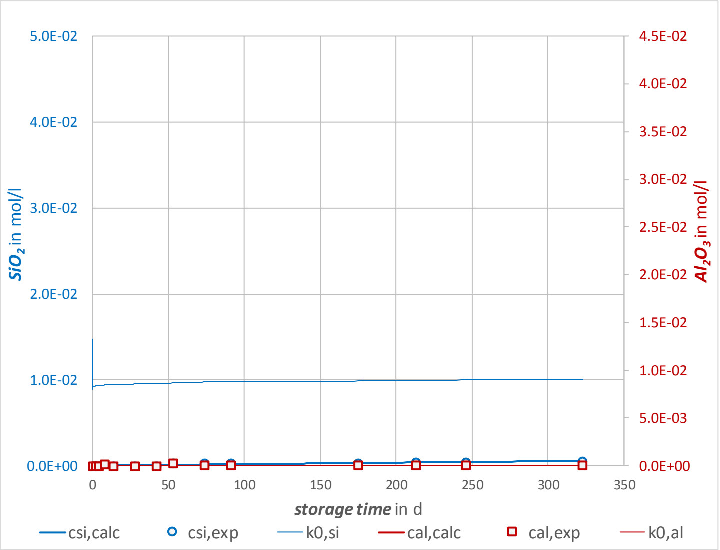

Figure 3.3: result of the calculation for 40°C and 0.1 M KOH

The important parameters can be summarized as follows:

$A_{Si}(t=0) = 0.21\ m^2/g$,

$k_{1,Si} = 4.79 \cdot 10^{-3}\ d^{-1}$,

$c_{Si,eq} = 1.7 \cdot 10^{-2}$ mol/l (for rock crystal in 0.1 M KOH, 40°C)

The diagram indicates that the solubility of quartz is lower under this condition in comparison to the dissolution conditions at 80°C. This can be measured and can be calculated too.

The diagrams show the Al_2O_3 concentrations too, which do not have any importance for the investigations at this time. However, the evaluation for the released alumina concentration is already prepared for aggregates, which provide not only silica but also alumina into the alkaline solution.

3.2 Calculation for quartzite

The same calculations were made with quartzite, here with different grain sizes according to the CPT (2-8 and 8-16 mm) in the solution system 80°C and 0.1 M KOH.

The assumption is that the thermodynamic calculated equilibrium concentration of the rock crystal can be transferred to quartzite. The very small muscovite amount is not considered.

Starting parameters for the calculation are:

$A_{Si}(t=0) = 1.71\ m^2/g$ for 2-8mm, and $1.64\ m^2/g$ for 8-16mm, determined by BET,

$c_{Si,eq} = 1.1 \cdot 10^{-1}$ mol/l (set equal to rock crystal for 0.1M KOH and 80°C).

Figure 3.4 demonstrates the results. The left the diagram shows the calculation curve for 100% quartz content. The measured solubility data are significantly higher than the calculated values. The best adjustment succeeds for a modified composition of the quartzite material, which means that the rock consists of approx. 75 wt.-% well-crystallized quartz and 25 wt.-% amorphous phases. Quartzite consists of strained areas and, obviously, such zones have to be considered in the calculation.

Here, there are now two parameters with which the calculated curves have to be adapted to the measured values, first the velocity constant $k_{1,Si}$ and second the amount of amorphous phases, expressed via the thermodynamic equilibrium data.

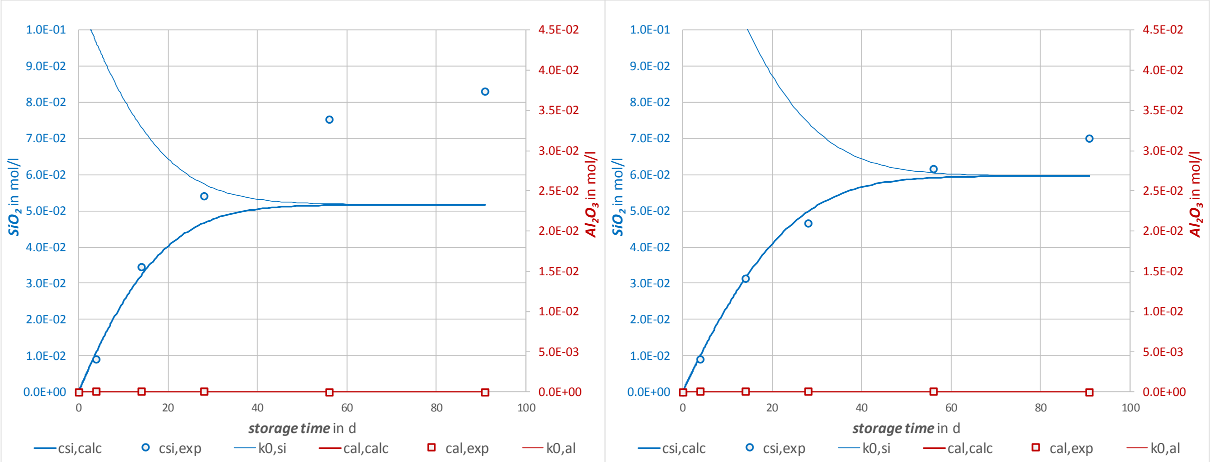

Figure 3.4: calculation for quartzite 2-8 mm, left 100%quartz, right 75% quartz, rest “amorphous”

The parameter based on the calculation are now:

$A_{Si}(t=0) = 1.71\ m^2/g$ for 2-8 mm, and $1.64\ m^2/g$ for 8-16 mm, determined by BET,

$k_{1,Si} = 6.39 \cdot 10^{-1}\ d^{-1}$ (2-8 mm); $3.87 \cdot 10^{-1}\ d^{-1}$ (8-16 mm),

$c_{Si,eq} = 2.5 \cdot 10^{-1}$ mol/l (2-8 mm); $2.4 \cdot 10^{-1}$ mol/l (8-16 mm).

The same effect can be measured with quartzite 8-16 mm (see Figure 3.5). The amount of strained quartzite areas can be specified in this case with 24%.

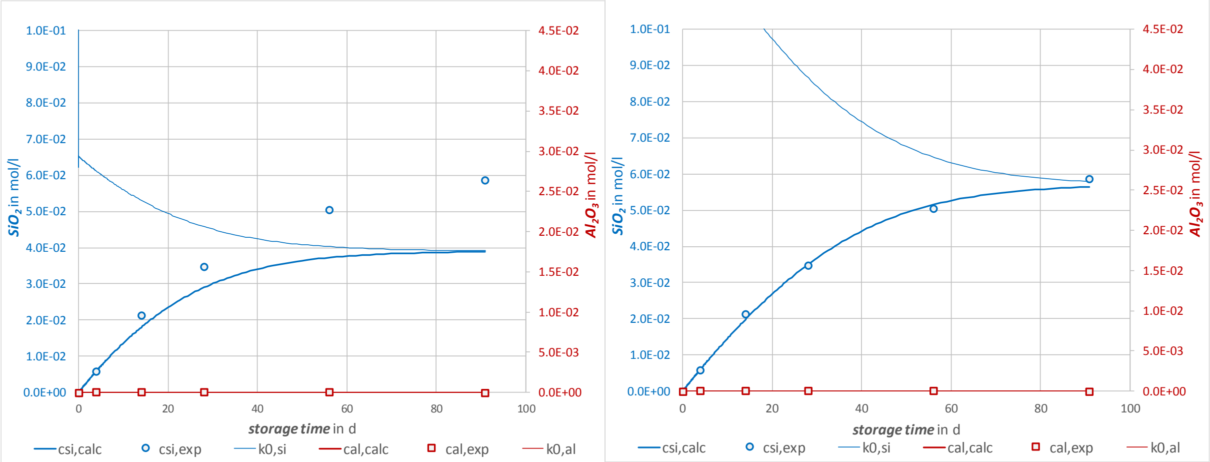

Figure 3.5: calculation for quartzite 8-16 mm, left 100%quartz, right 76% quartz, rest “amorphous”

3.3 Calculation for opal sandstone

The calculations were performed here also on the basis of the mineralogical composition in a first step. The quartz content (67wt.-%) and the amount of amorphous phases (14wt.-%) determine the value $c_{Si,eq}$. However, this calculation cannot be true. The result of the modified calculation, shown in Figure 3.6, bases now on 100% amorphous SiO_2. The explanation for this decision is demonstrated in Figure 3.7. Only the outer shale of an opal sandstone grain is dissolved by alkali attack, the nucleus consists of quartz and do not have any contact to the alkaline solution.

The parameter here are (for 0.1M KOH solution at 80°C):

$A_{Si}(t=0) = 1.34\ m^2/g$ for 4-8 mm, determined by BET,

$k_{1,Si} = 5.71*10^0\ d^{-1}$,

$c_{Si,eq} = 5.3*10^{-1}$ mol/l.

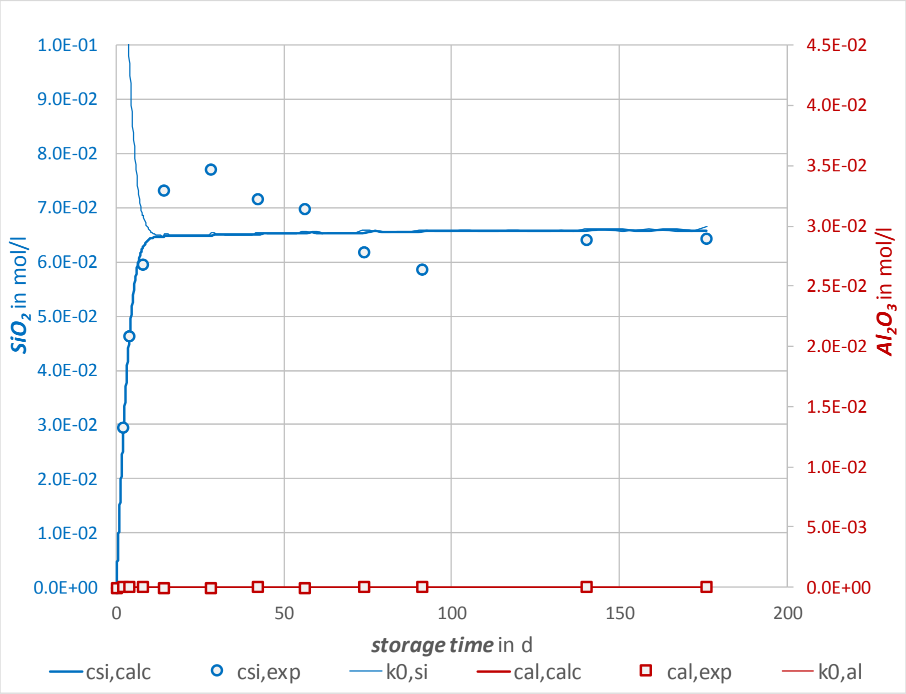

Figure 3.4: Calculation for opal sandstone 4-8mm (high density), conditions 0.1M, 80°C

Figure 3.5: reaction product of opal sandstone after alkali attack

3.4 Calculation for flint

The first calculations here were also made on the basis of the mineralogical composition. The quartz content (54wt.-%) and the amount of amorphous phases (46wt.-%) determine the value $c_{Si,eq}$. However, the calculation shows that the high amount of quartz does not provide any contribution to the solubility.

The parameter here are (also for 0.1M KOH solution at 80°C):

$A_{Si}(t=0) = 2.07\ m^2/g$ for 4-8 mm, determined by BET,

$k_{1,Si} = 1.03*10^0\ d^{-1}$,

However, the equilibrium concentration has to be adjusted to the measured values. The result is then:

$c_{Si,eq} = 5.3*10^{-1}$ mol/l (for amorphous SiO_2).

4. DISCUSSION AND CONCLUSIONS

Dissolution experiments with natural rock materials, which are used for the production of concrete, provide many different solubility results of such rocks. Because of the nature of rocks the composition, the structure, the porosity and so on can varied in a very wide range. The question is: It is possible to calculate the silica concentrations in alkaline solutions at different times and temperatures although only less parameters are known?

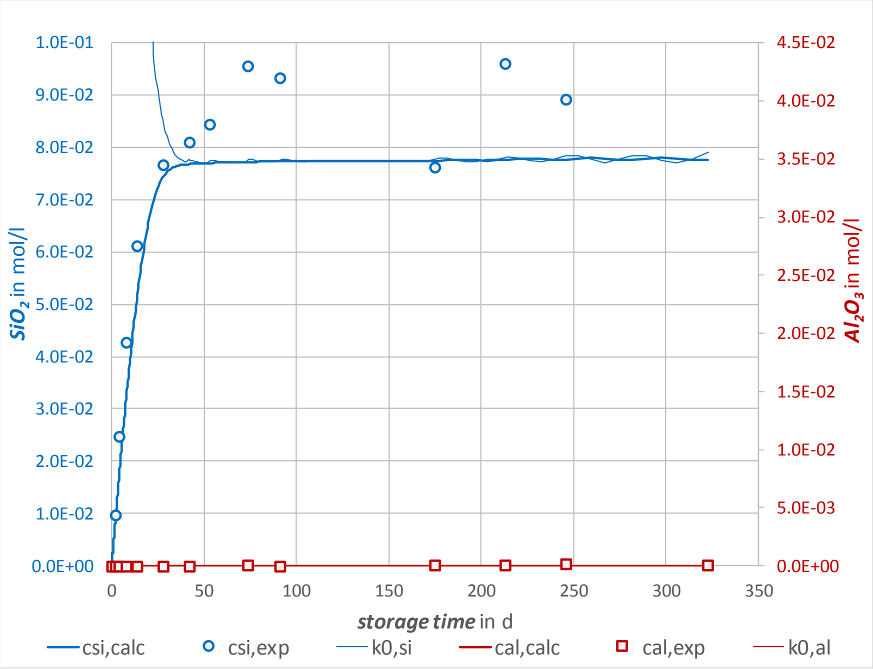

Figure 4.1: calculation for flint 4-8mm, conditions 0.1M and 80°C

To answer this question, a relatively simple kinetic model was developed and a basic thermodynamic data set were implemented. The kinetic model bases mainly on the surfaces and the surface changes of rock grains during dissolution. The surface of each grain has a very strong influence on the silica release. Calculations with only a little changes of the surface value provide a changed course of the calculated curve and the fit can be made not so well. The specific surfaces were determined by BET with nitrogen gas using a special measuring equipment. Maybe it would be better to use krypton gas because of the small surface values sometimes. However, the results of the calculations are satisfied and can very well interpreted. Important is to measure the surface of many original grains and to repeat the measurement three to five times.

The calculation results indicate that dissolution processes of natural aggregates in alkaline solutions are mainly controlled by the surface or surface properties of rock grains. It is not so important to have information on transport mechanisms inside of grains, more important are values, which can describe in detail the surface accessible from the outside.

The model bases too on the chemical and mineralogical composition of the rocks. In this case, the difficulties result of the possible differences between the surface composition and the average composition of the grain. It could be shown that, although flint has an average composition of 54wt.-% quartz and 46wt.-% amorphous phases obviously only the amorphous phase deliver a contribution to the silica dissolution.

Table 4.1: Thermodynamic data set for the system SiO_2 in water

	ΔH_{298}^0 kJ/mol	ΔG_{298}^0 kJ/mol	S_{298}^0 J/mol*K	C_{p298} J/mol*K	M g/mol
H_2O, aq	-285.83	-237.14	69.95	75.32	18.015
OH^-, aq	-230.02	-157.22	-10.90	-133.60	17.008
$Si(OH)_4$, aq	-1461.19	-1309.23	180.77	202.90	96.115
$Si(OH)_3O^-$, aq	-1431.83	-1253.06	90.87	-12.51	95.107
$Si(OH)_2O_2^{2-}$, aq	-1386.19	-1176.77	-11.93	-80.00	94.099
SiO_2, β-quartz, cr	-909.69	-855.62	42.63	44.67	60.080

Of course, the model bases also on thermodynamic data from the literature. In the last years, many values were published for such data of minerals. The model includes necessary data to calculate equilibrium concentrations for different minerals in dependence on temperature chosen for the experiments. For this an own database was created which includes certain mineral species.

Table 4.2: calculation of the equilibrium concentration for quartz

$SiO_2 + 2H_2O <-> H_4SiO_4$		
$0 = 1[H_4SiO_4] - 1[SiO_2] - 2[H_2O]$		
$\Delta_r H_{298}^0$ =	20.15	kJ/mol
ΔG_{298}^0 =	20.68	kJ/mol
$\Delta_r S_{298}^0$ =	0.00	kJ/mol*K
$\Delta_r C_{p298}$ =	0.01	kJ/mol*K
ΔH_T^0 =	20.57	kJ/mol
ΔS_T^0 =	0.00	kJ/mol*K
ΔG_T^0 =	20.74	kJ/mol
k_{Sol} =	8.57E-04	mol/l
c_{Si} =	1.16E-01	mol/l

Analogous calculations can be made with:

$Si(OH)_4 + OH^- <-> Si(OH)_3{*}O^- + H_2O$ and
$Si(OH)_3{*}O^- + OH^- <-> Si(OH)_2{*}O_2^{-2} + H_2O$.

An important result is the calculation of the equilibrium concentrations of SiO_2 for quartz, cristobalite and amorphous silica valid for different temperatures.

Values for 80°C are:

$c_{Si,eq}$ = 627 mmol/l (for 100% amorphous material),
$c_{Si,eq}$ = 116 mmol/l (for 100% crystalline quartz) and
$c_{Si,eq}$ = 267 mmol/l (for 100% cristobalite).
The same values for 40°C can be specified with:
$c_{Si,eq}$ = 120 mmol/l (for 100% amorphous material),
$c_{Si,eq}$ = 17 mmol/l (for 100% crystalline quartz) and
$c_{Si,eq}$ = 44 mmol/l (for 100% cristobalite).

Ultimately it is important, that the model bases on kinetic equations, which consider the changes of the surface with every calculation step (with every changes of silica in the solution) and the changes of the OH^- concentration during the dissolution process. Of course, each calculation step was performed by consideration of charge balancing. The limiting factors to calculate dissolution curves in the model are in general:

- the mass and therefore the surface of the material (important for rocks, more important for powdery materials – SCM -, which react very fast),
- the equilibrium concentration of silica under certain conditions (for rock crystal the equilibrium is not reached because of the very small surface, for flint and opal sandstone the equilibrium state is almost reached, quartzite comes close to the equilibrium limit under the conditions chosen).).

Even if there are still some unanswered questions, the relatively simple model already provides satisfactory results for the description of the dissolution processes of aggregates in highly alkaline solutions. The prediction of the solubility behaviour of aggregates over a long period is just as possible as the prediction for different pH values. More or less unknown are the velocity constants of the dissolution. It can be concluded, that only a few measurement points at the beginning of the dissolution experiments are necessary to find the slope of the curve (maybe until 10 to 14 days or even less). The further course of the curve then can be calculated.

Because knowledge of the solubility behaviour of aggregates is an important requirement for expiry of a harmful ASR, an important step was done for understanding reaction mechanism of aggregates in highly alkaline solutions.

The model should be transferred to more complex composed grains, which consist of different minerals. The model can also be used to understand, maybe in a better way, the dissolution behaviour of SCM`s of course in combination with aggregates.

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