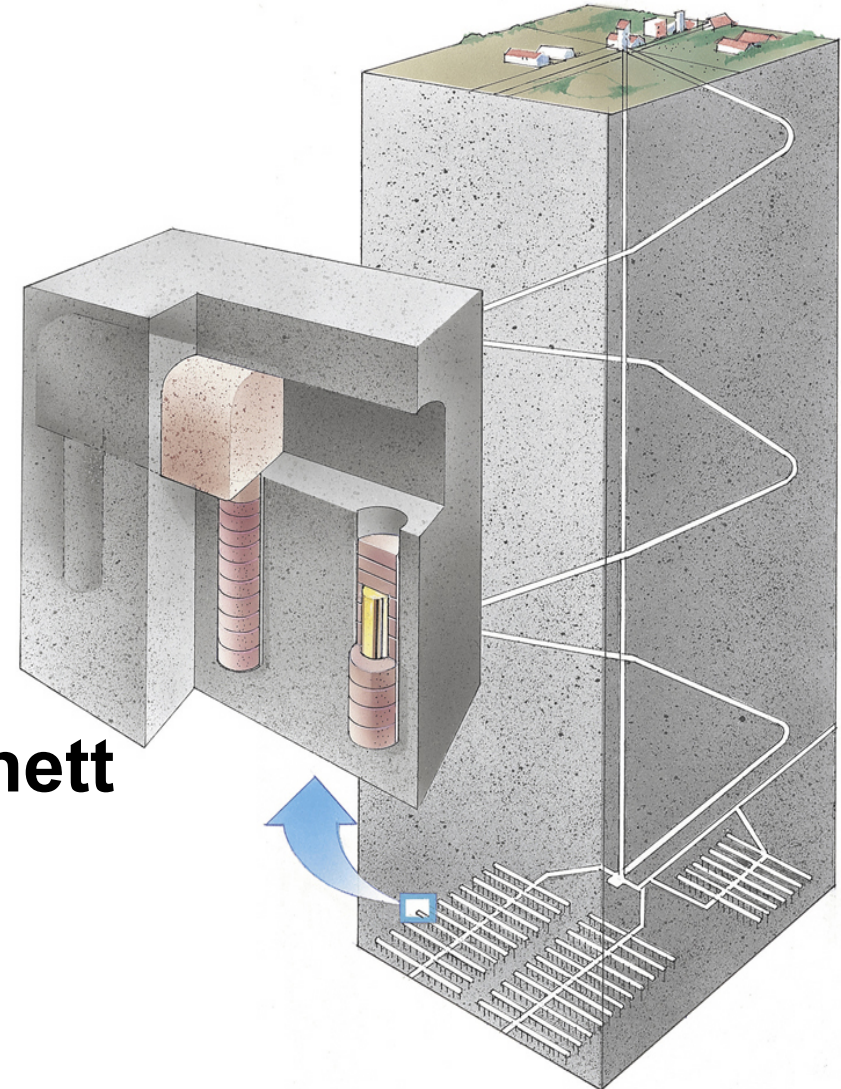
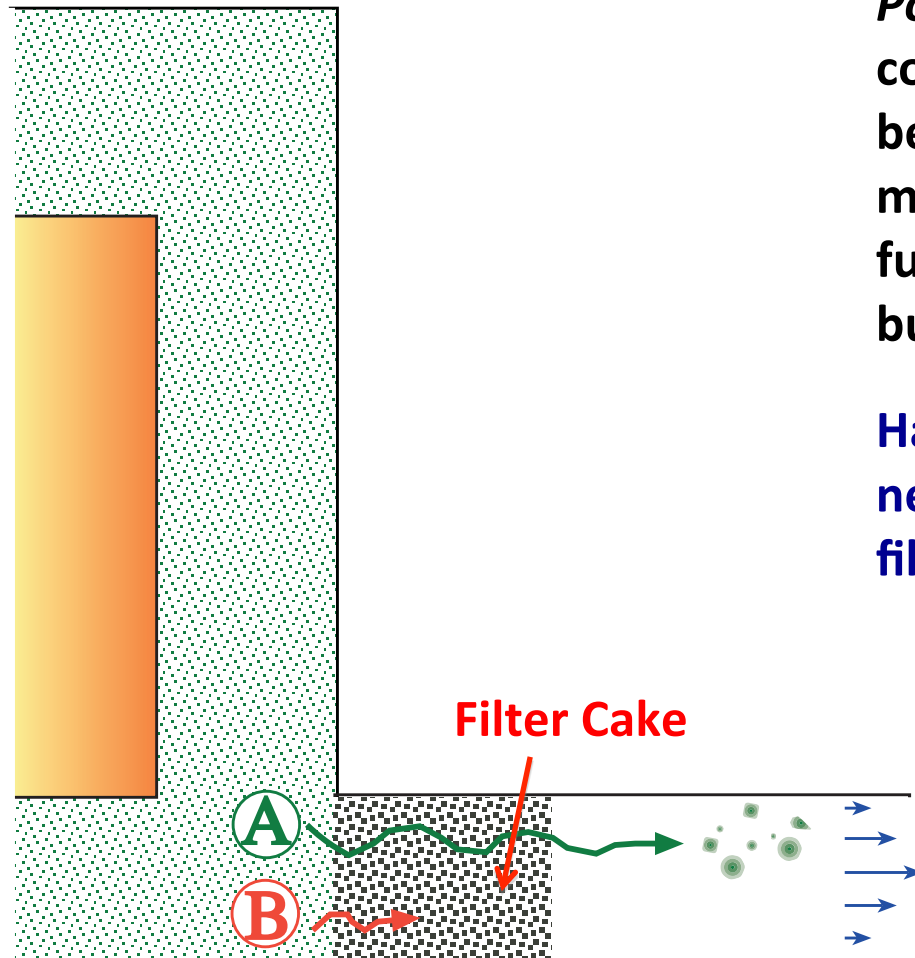


Buffer Erosion: Filtration *A Decision-Tree Approach*

**Mick Apted and David Bennett
BRITE**



Filtration Mitigation of Buffer Erosion

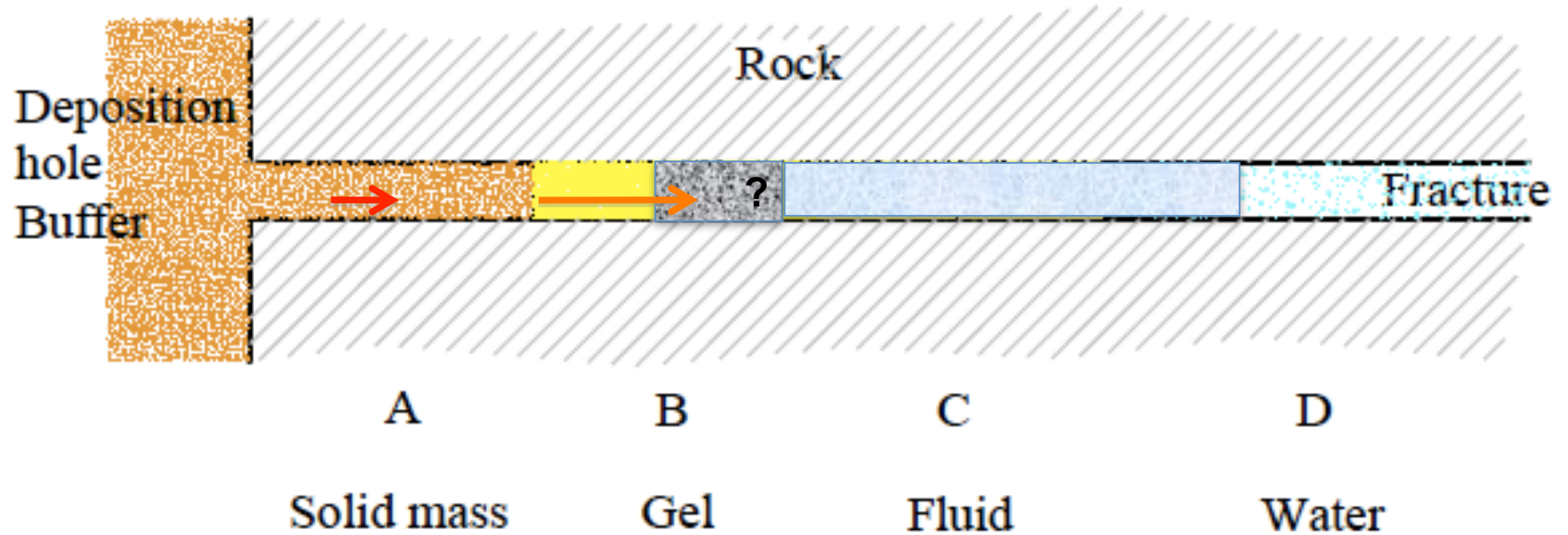


Postulated Scenario: Removal of clay colloids from extruded buffer leaves behind a ‘filter-cake’ of accessory minerals that could prevent by filtration further release of clay colloids from the buffer.

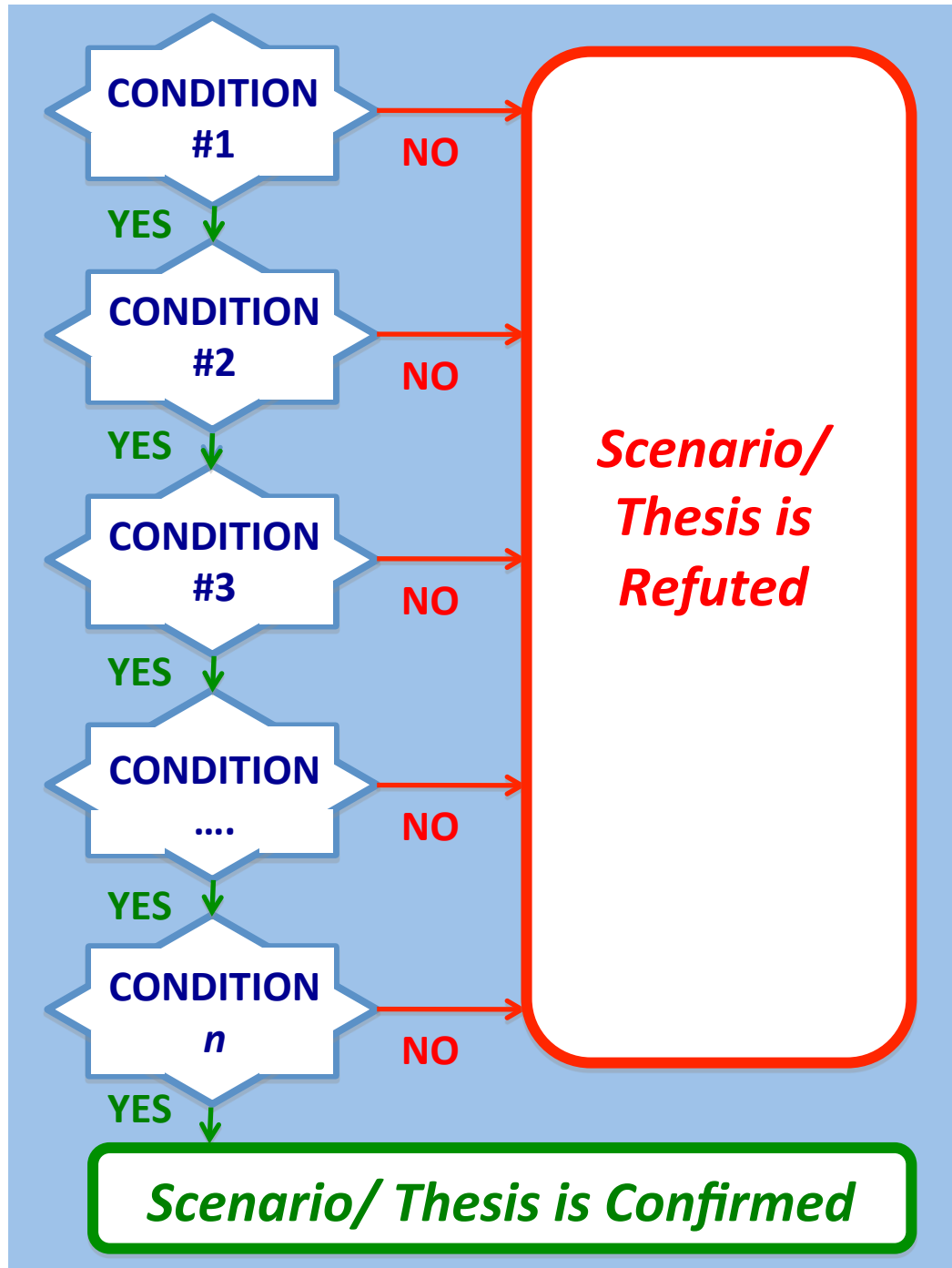
Has SKB obtained information and analyses necessary to establish the reliability of filtration to mitigate buffer erosion?

- T. Richards (2008) Particle Clogging in Porous Media, Chalmers University [REFERENCE 1]
- Birgersson et al., (2009) “Chapter 4 Filtration” in Bentonite Erosion, Clay Technology, Lund [REFERENCE 2]

Evolution of Filter Cake



Decision Tree Approach



- Speculative scenarios typically involve multiple lines of evidence and arguments to support their basis.
- ALL of the Conditions must be “YES” to reliably confirm.
- It only takes one “NO” to refute.

Conditions for Filtration

- **Is Filtration Theory Valid for Buffer-Repository Environmental Conditions?**
- **Do “Filter Cakes” of Non-clay Minerals in Buffer Form under Repository-Relevant Environmental Conditions?**
- **Are “Filter Cakes” Effective in Mitigating Further Loss of Clay Colloids?**
- **Do “Filter Cakes” Persist over Relevant Repository Timescales and Future Events?**
- **Can a Dependable Safety Function be Assigned to the Characteristics of a Degraded Barrier?**

Is Filtration Theory Valid for Buffer-Repository Environmental Conditions?

- **Multiple theories developed and tested in industrial contexts.**
 - “DLVO and the convective-diffusion equation predicts the right magnitude of colloid removal efficiency when no energy barrier exists; however, when energy barriers exist, discrepancy is large.”
 - **Packed-bed approximation seems favored [REF 1]**
- **Limitations in Applying Any of the Theories**
 - Charged (energy) surfaces, multiple forces
 - Presence of fluid phase in addition to assumed 2 ‘surfaces’
 - Irregular shape, size and distribution of solids in realistic filter-cake
 - Change in filter porosity (‘clogging’)
 - Many empirical (‘fitting’) factors, hence, limited ability to extrapolate results to different bentonites (or variations in any single deposit)
- **Evaluation: “YES”, but important limitations**

Packed Bed Schematic

➤ Theory Limitations

➤ Particle Shape:

spheres vs. irregular

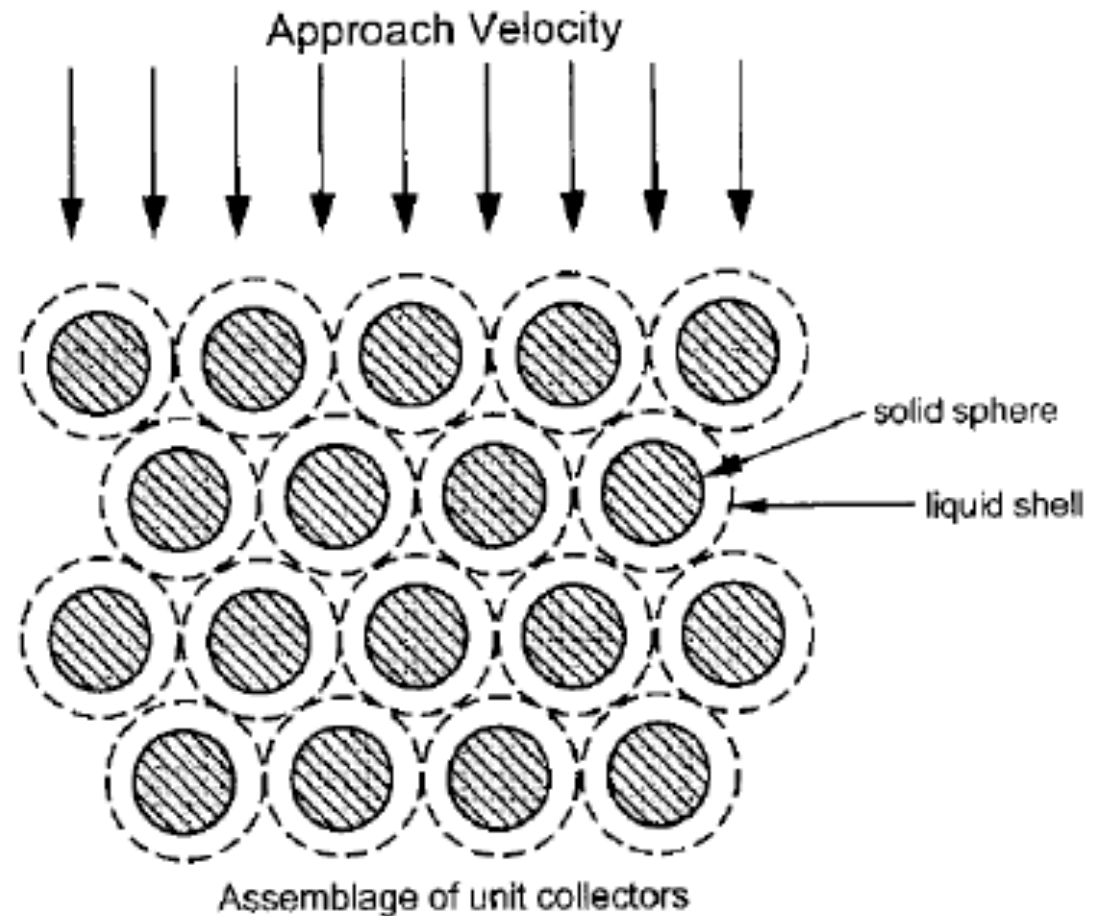
➤ Particle Size:

uniform diameter vs. distribution of sizes

➤ **Geometry:** Closest packing, minimal porosity

➤ **Pore Shape and Sizes:** uniform and regular vs. distributed and irregular.

➤ **Flow:** How can water flow out from buffer into filter cake?



Do “Filter Cakes” of Non-clay Minerals in Buffer Form under Repository-Relevant Conditions?

- Formation of a ‘filter-cake’ under repository conditions, as buffer extrudes and erodes into fractures, seems to be simply **assumed** based on observed preferential loss of clay-colloids that leaves behind a ‘**collection of accessory minerals**’; quartz, feldspar, pyrite, oxides, organics(?),...
- However, such a ‘**collection of particles**’ may not be the same as the uniform ‘**packed filter-cakes**’ or **metal filters** used in REF1 and REF 2 experiments, respectively.
- No post-experiment analysis to confirm **development and properties of realistic ‘filter-cake’** from buffer erosion [REF 2 simulated development, but no post-test analysis of solids].
- No **natural analogues** cited or identified as a useful area of confirmatory study.
- Evaluation: “**NO**” (at least no confirmation of representativeness of uniform ‘**packed bed**’ or **metal-filter** analogues)

Accessory Minerals Preparation [REF 1]

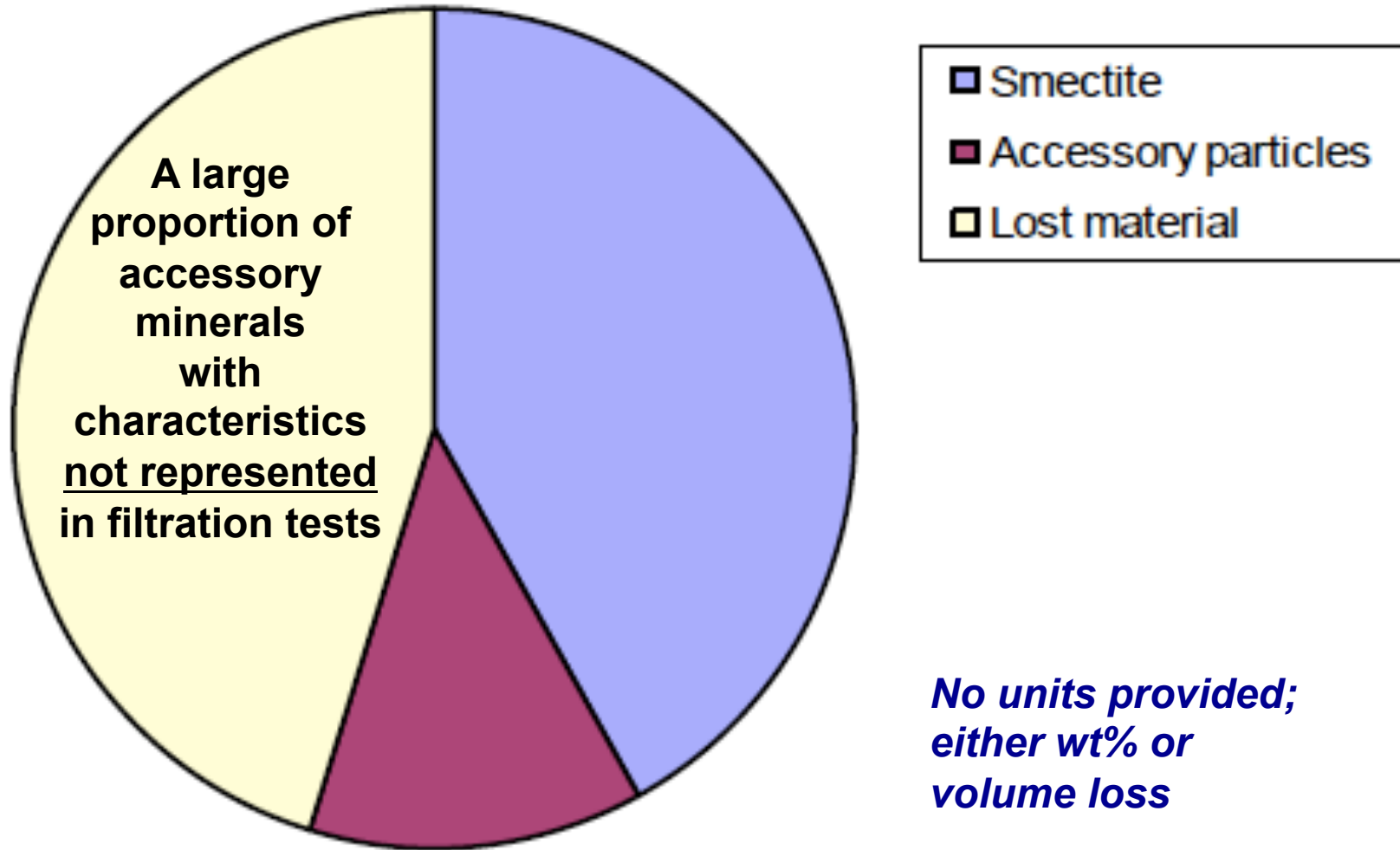
- MX-80 treated with 1 M NaCl to ion-exchange all sites.
- Several washings and centrifugation steps to remove suspended clay-gel and obtain 'accessory minerals'
- 'Collection of minerals' = 'Packed bed' filter-cake or loose slurry?



Are “Filter Cakes” Effective in Mitigating Further Loss of Clay Colloids?

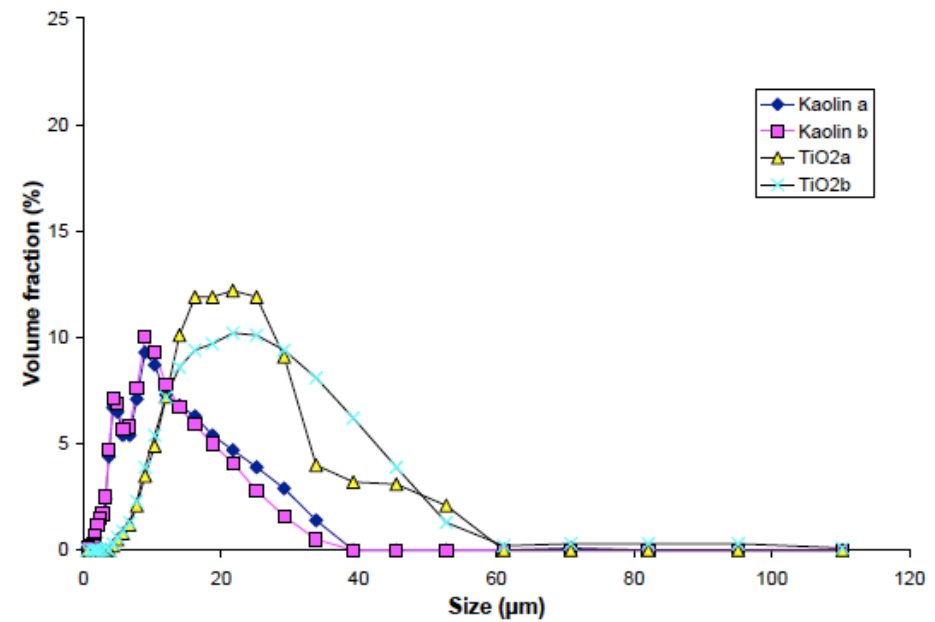
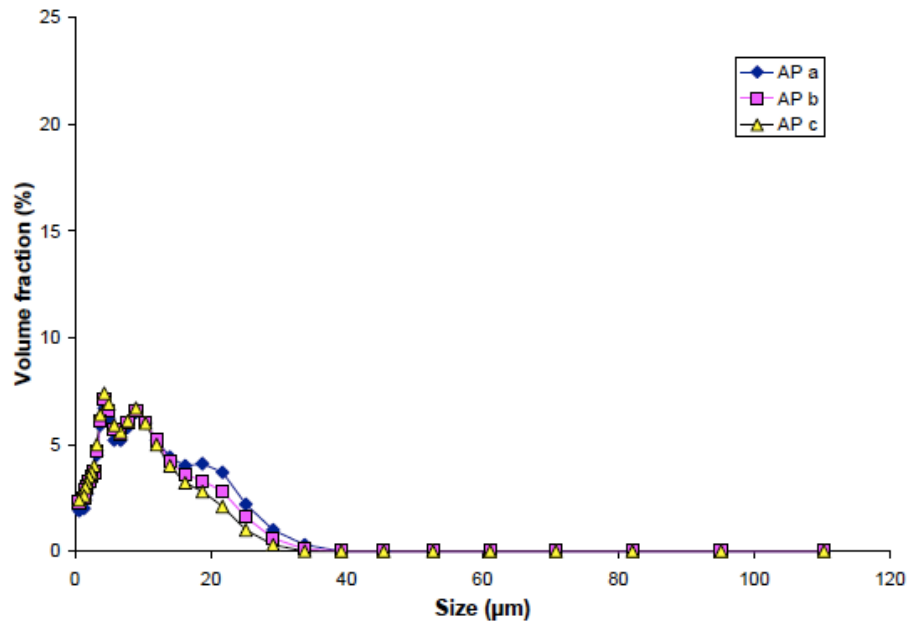
- **REF 1: Pressure-induced flow of suspension of 200-nm diameter smectite through pre-formed ‘filter cakes’ of Na-MX80 or additives**
 - 5-10 μm diameter accessory minerals
 - different ratios of industrial materials (potential ‘additives’)
 - Sensitivity tests: thickness of filter cake and pH effects
- **REF 2: Diffusion of clay-colloids, formed within various buffer ‘compositions’, through various size and thickness pre-formed filters into 1 ml/min circulating deionized water**
 - MX-80, homoionic Na- and Ca-montmorillonites, kaolin and diatom additives, ‘ $> 2 \mu\text{m}$ ‘accessory minerals’ from MX-80
 - Filtration effectiveness measured by (1) swelling pressure drop in ‘buffer’ sample, and (2) turbidity measurements
 - Attempt to ‘grow’ a filter cake from MX-80

Loss of Solids during Preparation [REF 1]



Particle Size and Shape [REF 1]

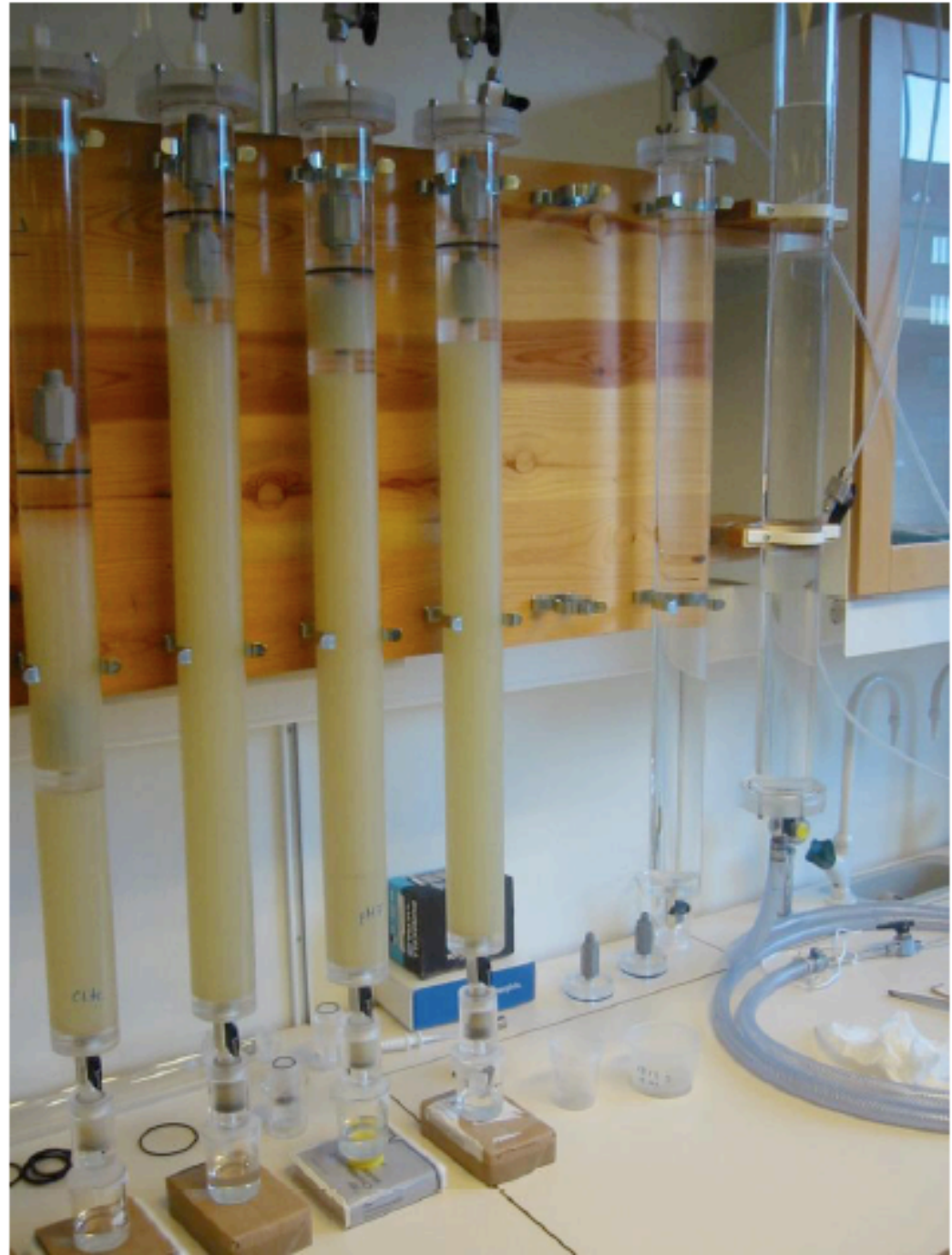
- “It must be emphasized that the measured particle size is a conversion from the actual geometry to a spherical geometry and the particles may have another shape that significantly differs from a sphere.”



Test Device

[REF 1]

- 5-bar pressure-induced flow (?) of suspended clay colloids through packed-bed filter cakes
- All columns controlled by same pump; flow hysteresis during column exchanges acknowledged
- No analysis of possible non-filtration factors (e.g., colloid stability) affecting measured filtration efficiency



Estimate Filtration Coefficient (1)

➤ $\ln\left(\frac{C}{C_0}\right) = -\frac{3}{4}(1-\varepsilon)\alpha\eta\frac{L}{a_c}$ Eq.(24)

- where C is the concentration of suspended clay particles at the end of the cake, C_0 is the incoming concentration, ε is porosity, a_c is the radius of a spherical collector of the filter cake, L is the filter cake length, is the empirical collision frequency (0.001 to 1), and η is the efficiency of a single spherical collector.

➤ $\eta = \frac{I}{UC_0\pi a_c^2}$

- where U is the flow velocity and I is the deposition rate.
- By comparing the concentration outside the filter cake with C obtained from Eq. (24), estimate the deposition rate. Or by estimating the deposition rate from the slope of 'Filtrate vs. Time' (Figure 11), compare concentrations.

Estimate Filtration Coefficient (2)

- Figure 11 [REF 1] compares different sample preparations and thicknesses
- “It is clear that the more material in the cake the less filtrate will penetrate.” (this conclusion does not seem to be fully supported by the data)

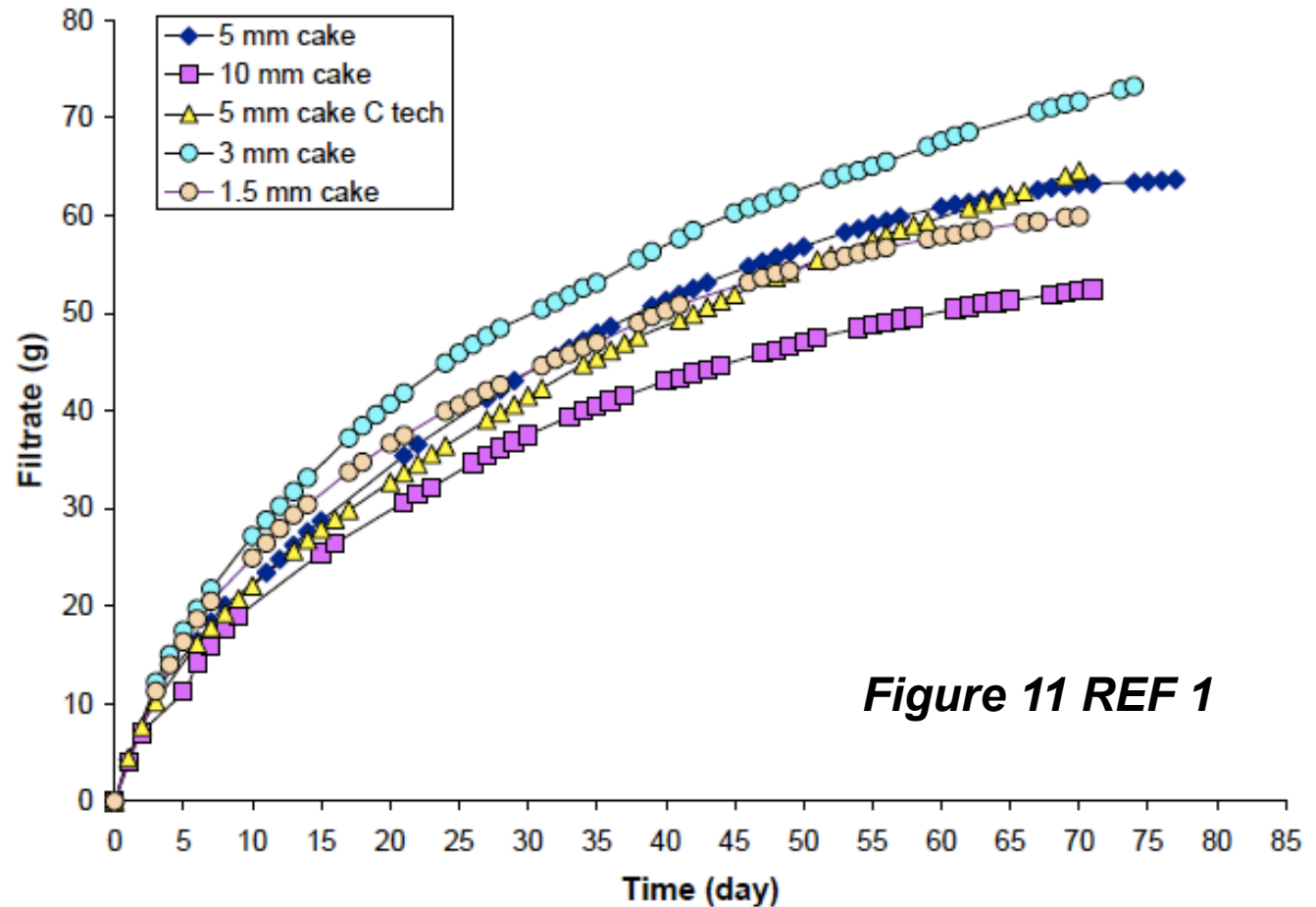


Figure 11 REF 1

Estimate Filtration Coefficient (3)

- The purpose of the comparison is to help estimate the filtration coefficient, Λ , which could be useful in modeling of the filtration process using a more familiar form of the mass transfer equation:

- $$\frac{dC}{dt} = -\Lambda C + D\nabla^2 C - \nabla \cdot (\mathbf{U}C)$$

- where C is the concentration of suspended clay particles, first term on RHS is the filtration term, the second term on the RHS is the diffusion term, and the last term on RHS is the advection term.

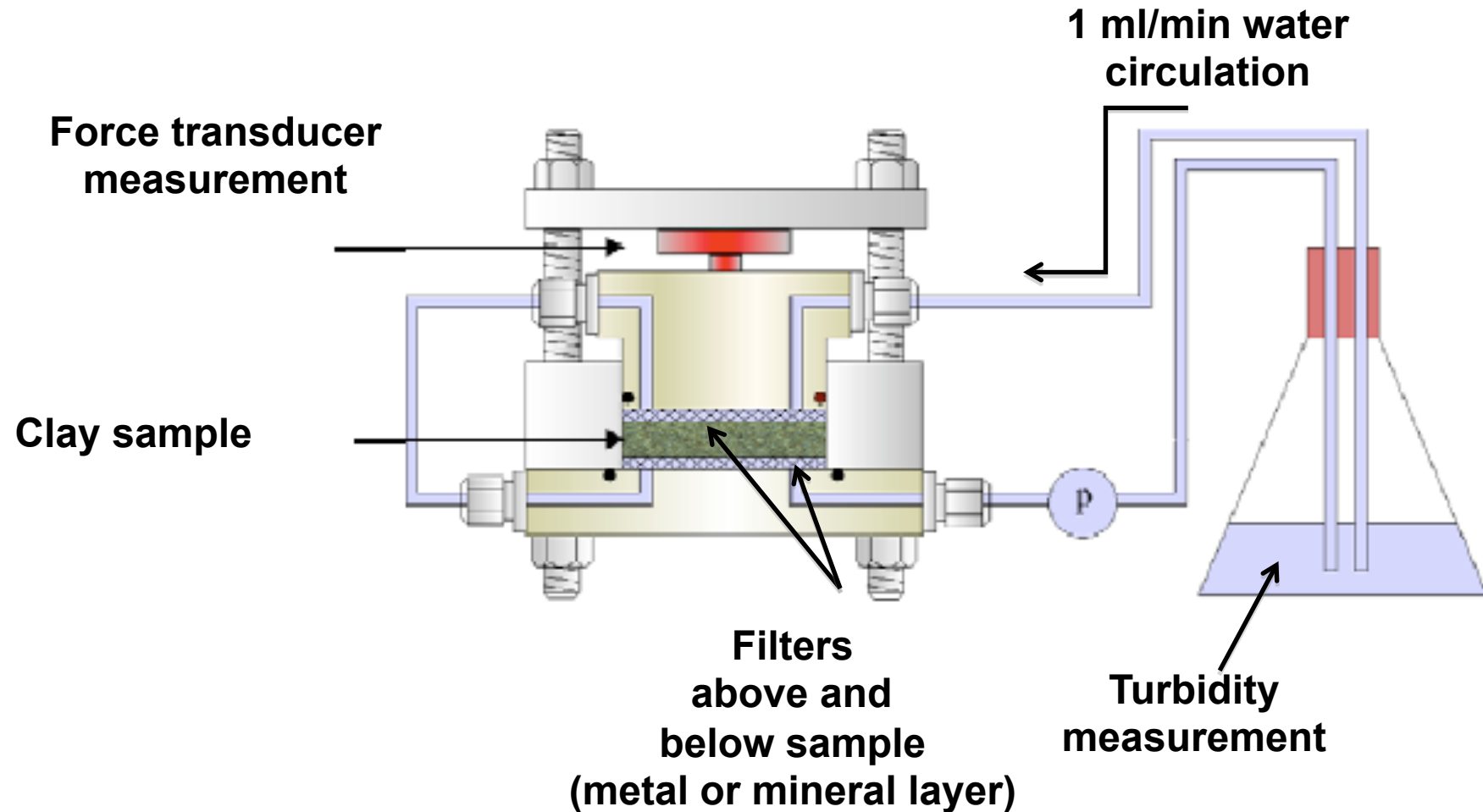
Conclusions [REF 1]

- “The amount of material in the filter cake has an impact of the filtration rate. More material decreased the filtration rate. [Not always]
- The separation of smectite particles was almost complete in all tested cases. Only a fraction of the solid concentration in the suspension was found in the filtrate.
- The amount of solid particles in the filtrate and the filtration rates were comparable for all different pH levels (no pH influence on clogging).
- Gravitation may influence the filtration rate. However, the separation quality is equally good regardless of upward or downward filtration.
- It is possible to achieve a good separation of smectite particles from a water suspension with a mixture of additive materials. The one most promising seems to be kaolin, which of the investigated materials, most resembles the properties of the accessory material particles.”

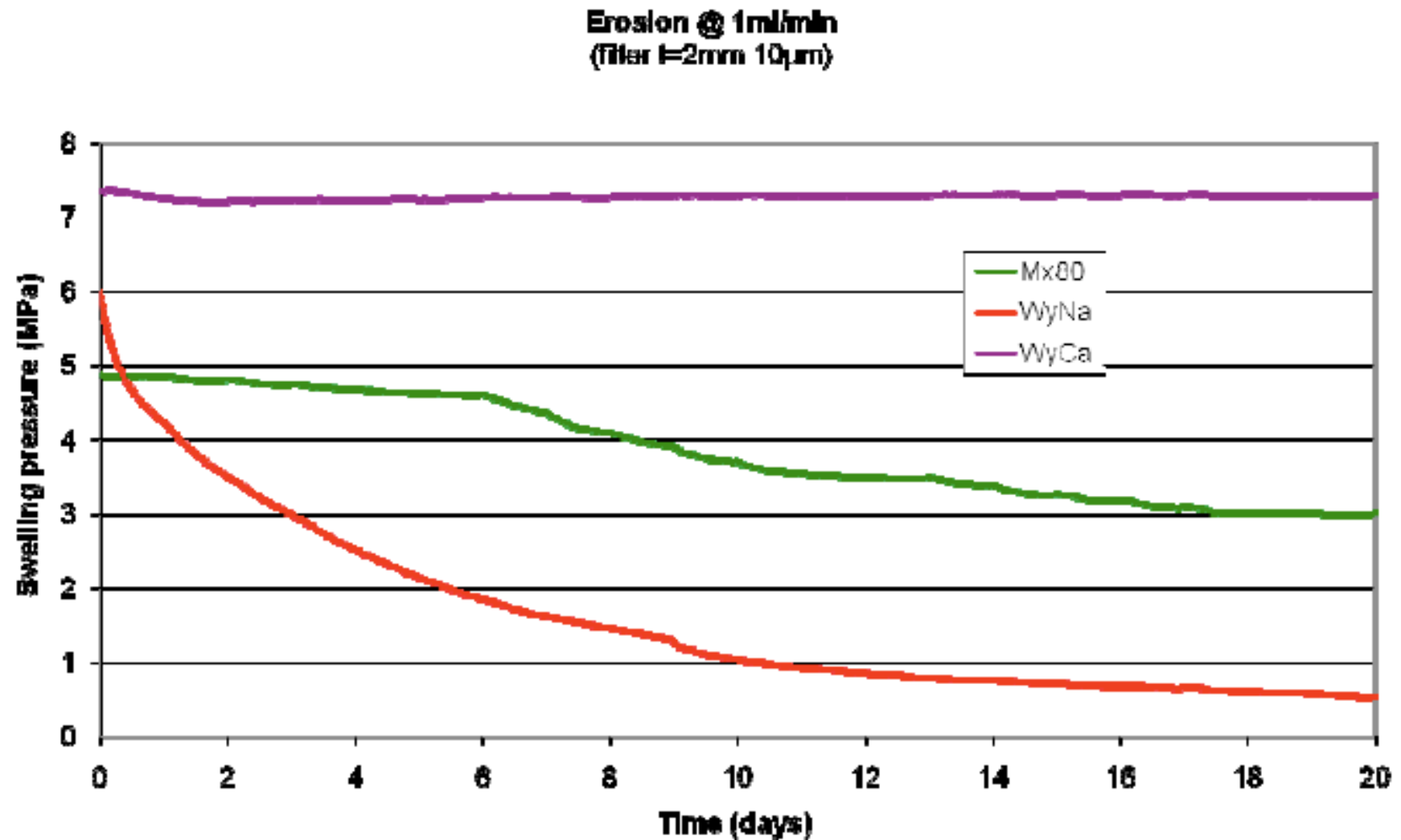
Filtration Tests [REF 2]

- **What physical barrier can stop colloid migration by filtration?**
- **Materials tested**
 - Untreated MX-80
 - Homoionic Na-form and Ca-form montmorillonite
 - MX-80 accessory minerals (> 2 μm in diameter)
 - Additives: kaolinite [$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$] and diatomite [SiO_2]
- **Test conditions for measurement of filtration efficiency**
 - 1 ml/ min circulation of deionized water across filter
 - 2-mm thick metal filters of prescribed pore size: (0.5 to 100 μm)
 - Same initial density of clay sample = 2000 kg/m^3 (wet)
 - **MEASURE:** Change in swelling pressure of different clays in load cell
 - **MEASURE:** Turbidity change in circulated water outside of filters

Test Device [REF 2]

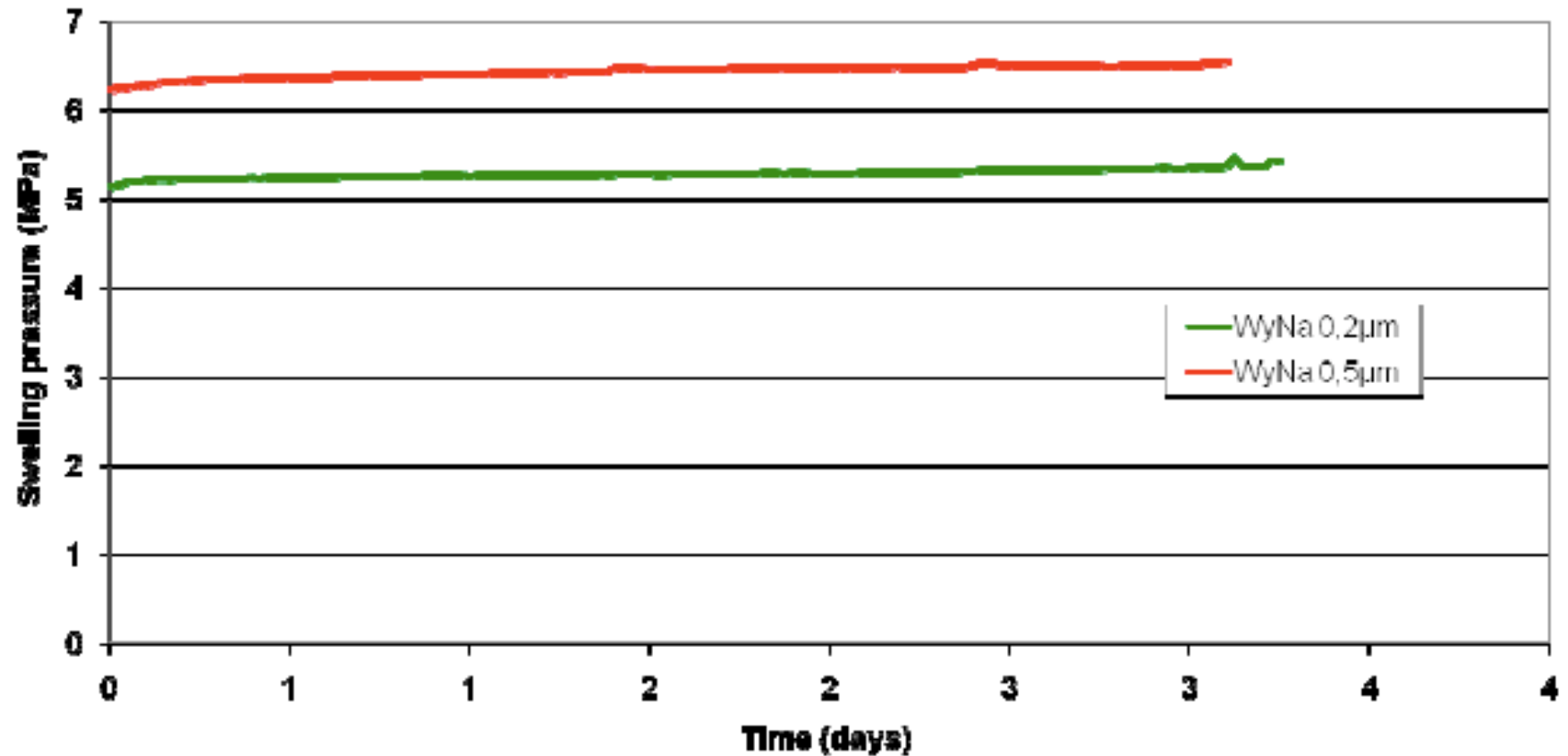


Comparison of Untreated MX-80 with Na-and Ca-montmorillonites



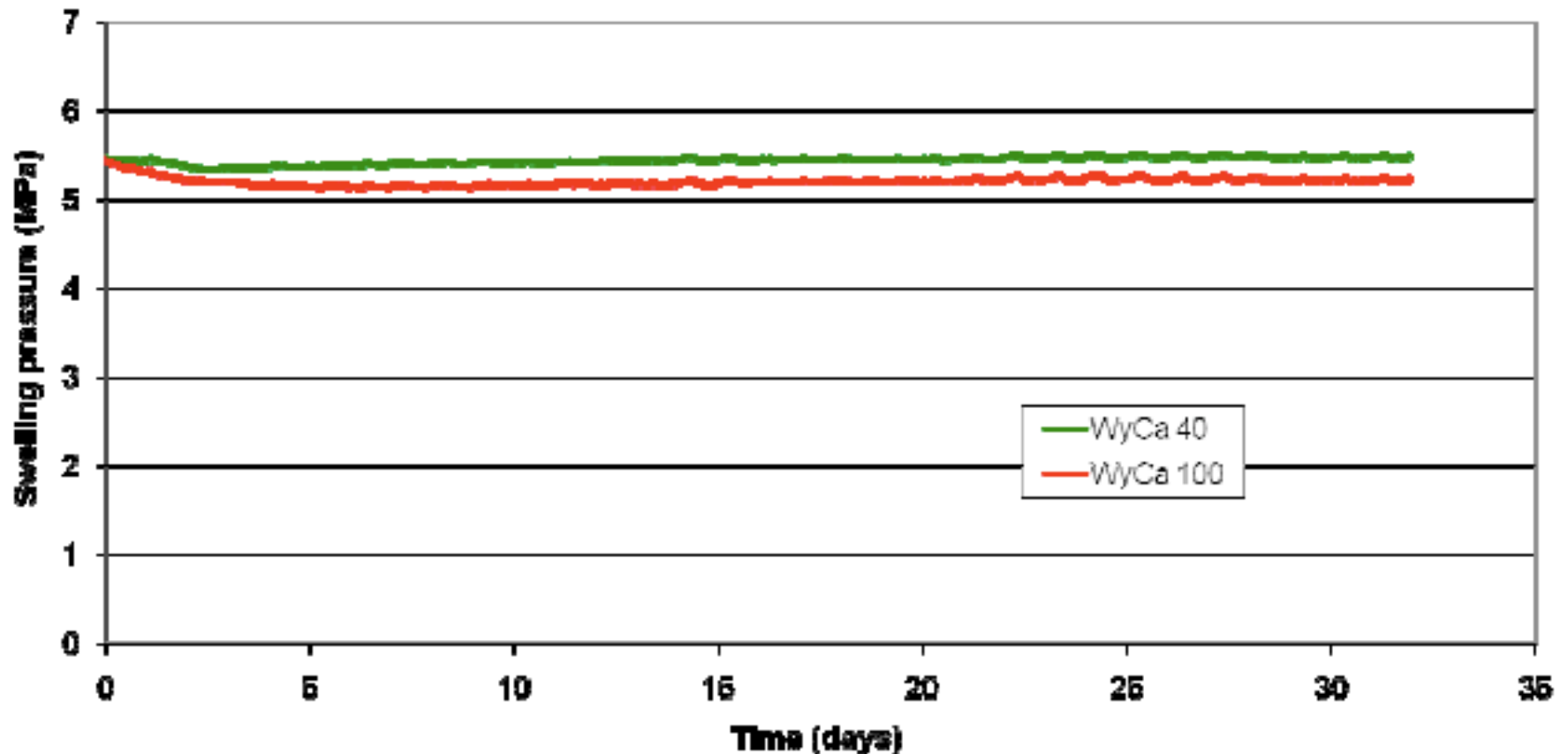
Lower-Bound Filter for Na-montmorillonite

Erosion @ 1ml/min
(filter t=2mm 0.2µm/0.5µm)



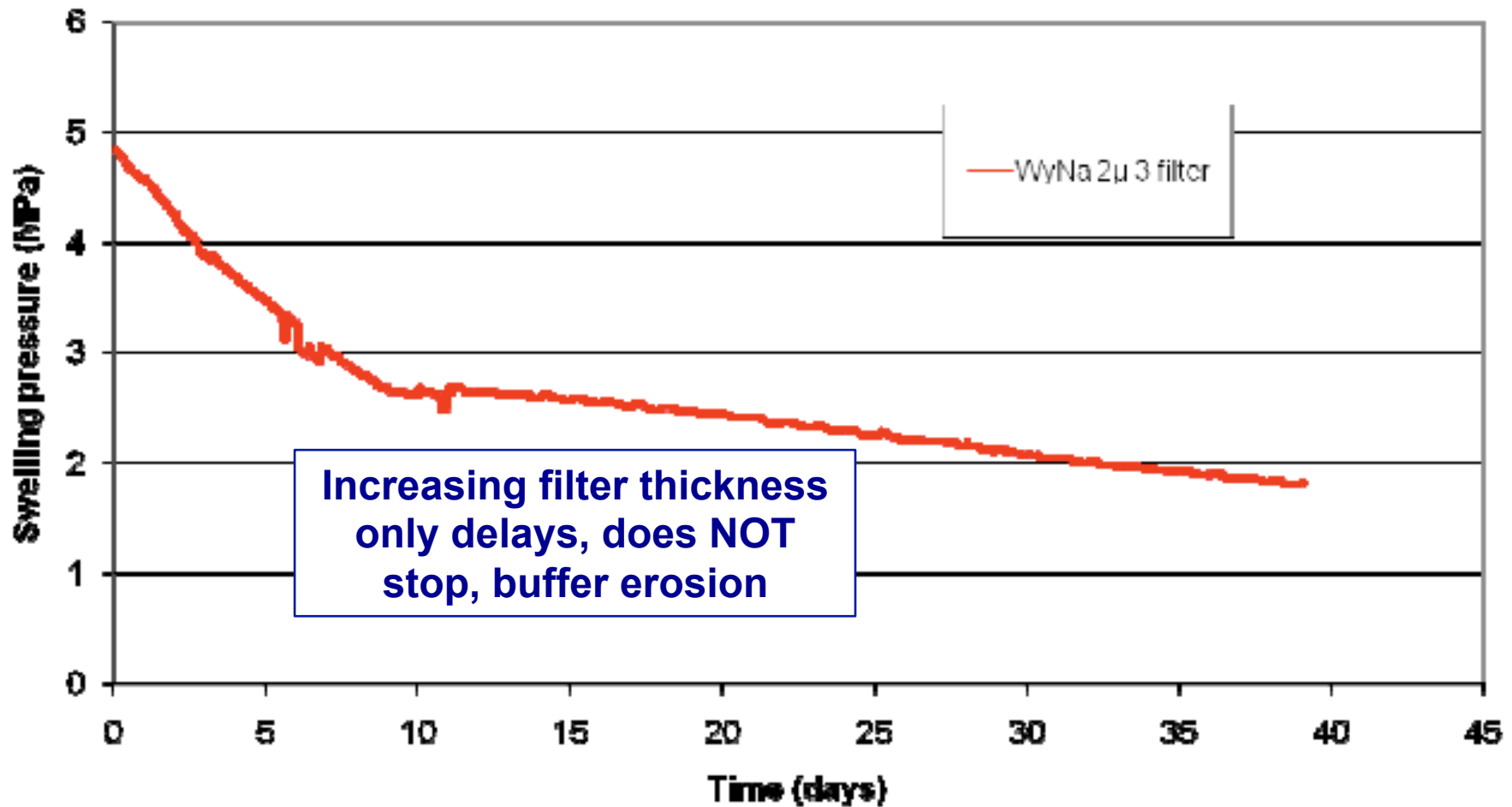
Upper-Bound Filter for Ca-montmorillonite

Erosion @ 1ml/min
(filter $t=2\text{mm } 40/100\mu\text{m}$)



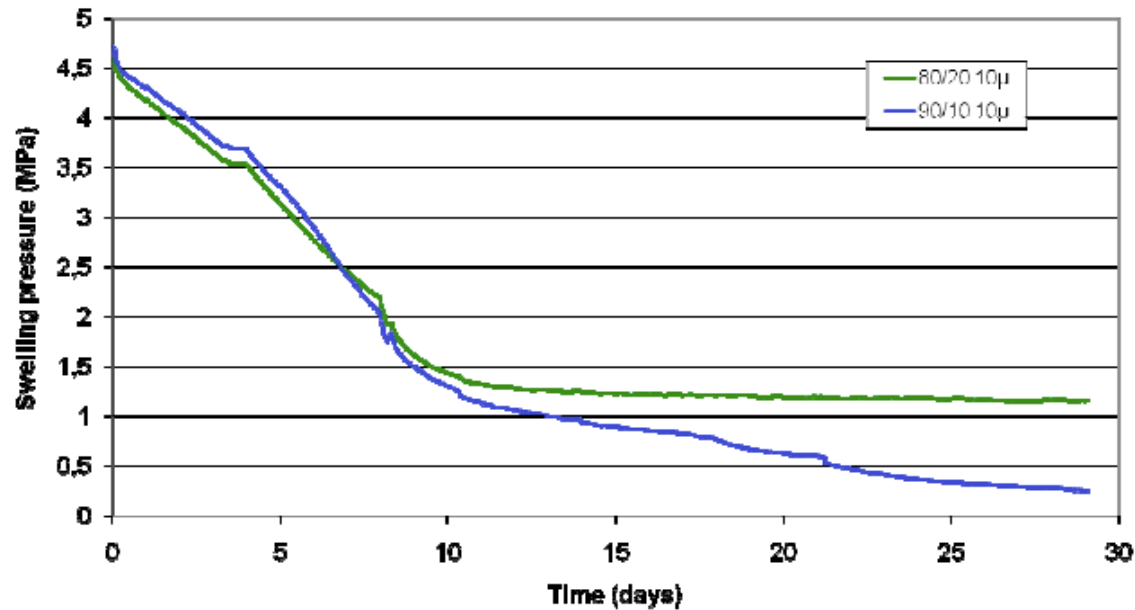
Effect of Filter Thickness

Erosion @ 1m³/min

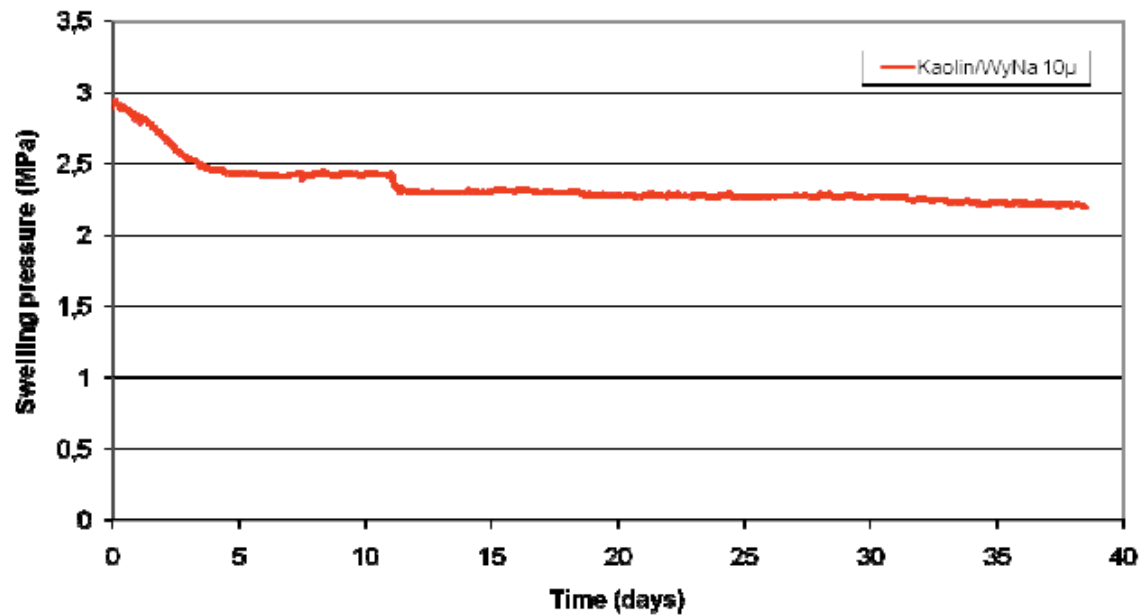


Effect of Kaolin Additive

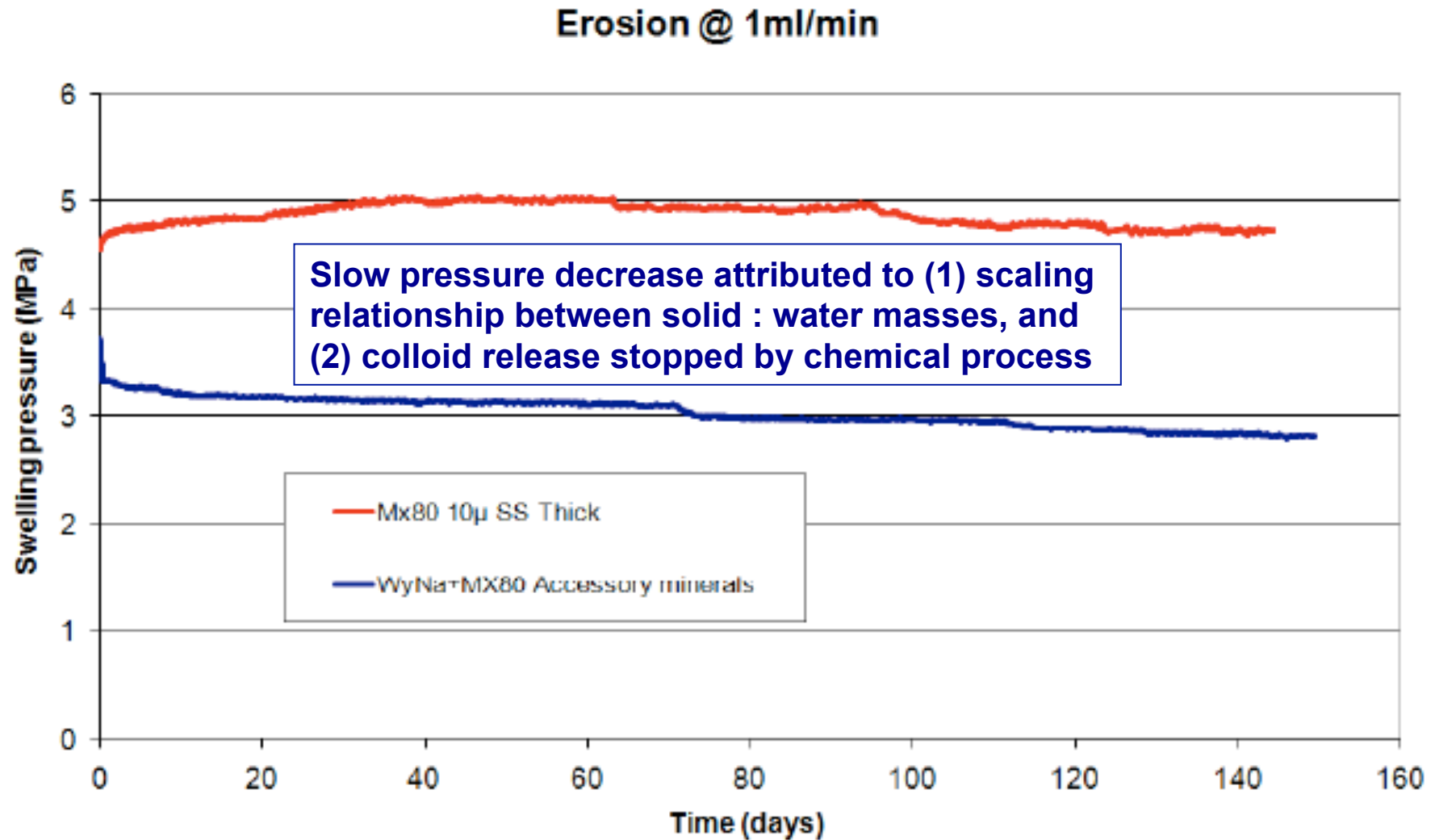
Erosion @ 1m/min



Erosion @ 1m/min



Growing an Accessory-Mineral Filter-cake



Conclusions [REF 2]

- **“A number of tests have been made with both untreated MX80 bentonite from Wyoming and homoionic sodium and calcium converted clays (montmorillonite). Combinations of these and filler materials such as kaolinite, diatomite and accessory minerals was tested. Also a number of filters with different pore size have been tested.”**
- **The following combinations have been found to stop colloid release into de-ionized water at a flow of 1 ml/min**
 - **Homoionic calcium clay and pore size < 100Lm.**
 - **Homoionic sodium clay and pore size < 0.5Lm.**
 - **Homoionic sodium clay and a compacted 2 mm layer of either kaolinite or diatomite.**
 - **Other filler materials can be expected to have similar sealing properties as kaolinite provided similar grain size distribution.”**
- **Missed opportunity to examine development and structure of a filter-cake of accessory minerals formed from buffer erosion of MX-80**

Are “Filter Cakes” Effective in Mitigating Further Loss of Clay Colloids?

➤ Concerns

- relevance of 1 bar test conditions for REF 1 and 2 tests
 - relevance of pressure flow vs. diffusion colloid transport in REF 1
 - assumed ‘spheres’ vs. irregular size, shape and distribution of particles for both REF 1 and 2 tests
 - Sample preparation: ‘gentle’ crushing leading to possible change in size and reactivity of surfaces
 - flow hysteresis during REF 1 tests
 - 55% of buffer material lost in preparation for REF 1 (unreported in REF 2)
 - no post-test microscopic analysis of solids for either REF 1 and 2
 - evidence of compression/ compaction/ clogging of filter cake that negates assumed theoretical assumptions
- **Evaluation:** “**YES**”, but concerns about range of relevance

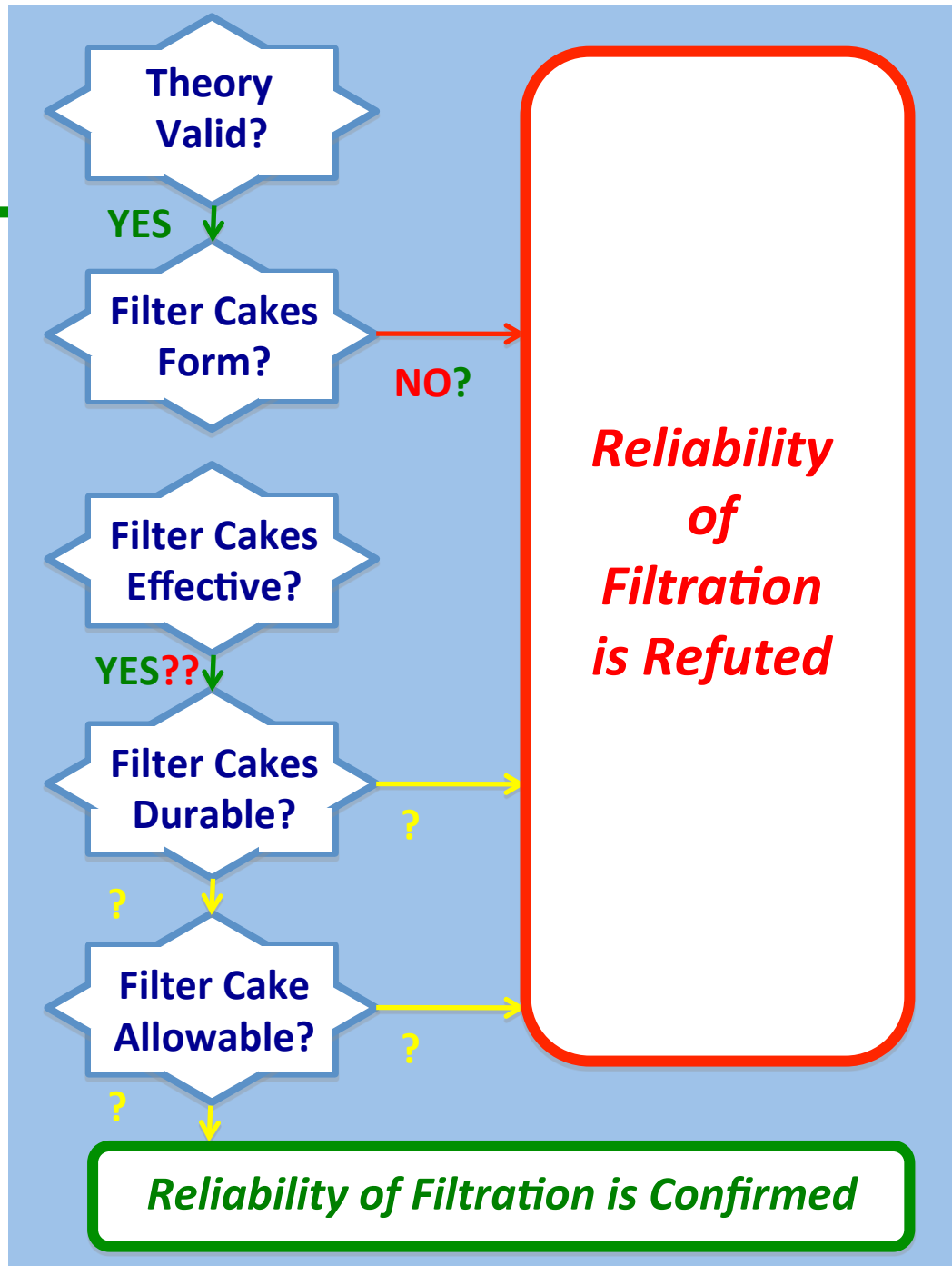
Do “Filter Cakes” Persist over Relevant Repository Timescales and Future Events?

- This question does not seem to have been explicitly considered.
- Concerns
 - If formed, will ‘filter cakes’ persist after
 - earth tremors
 - change in fracture-flow rate or direction
 - slight shearing or expansion/compression of fracture
 - For repository depths and timescales, is long-term pressure-solution of accessory minerals in filter-cake (brittleness) a possibility?
 - Natural analogues would be especially useful as support
- Evaluation: “**OPEN**”, no analysis presented

Can a Dependable Safety Function be Assigned to the Characteristics of a Degraded Barrier?

- This is a broader regulatory question.
- If non-QA' ed properties favorably affecting performance and safety can be assumed for a degraded barrier, why require QA of initial/ emplaced barriers?
- Can tests done on MX-80 be reliably extrapolated to other bentonites? Seems doubtful (empirical approach)
- Same question could arise in other areas
 - Mass-transfer resistance of failed copper canister
 - Limited surface exposure of cladded spent fuel
- Evaluation: “**OPEN**”, not yet considered (also by SSM)

Summary of Evaluation



- Theory and lab data provide limited support regarding filtration effectiveness for pre-formed MX-80 and some additives (does SKB plan to re-engineer buffer?)
- Tests have many concerns and limitations, however
- These MX-80 results may not extrapolate to other bentonites or later MX-80
- *Formation, durability and regulatory acceptability* of filter-cake mitigation of buffer erosion are not confirmed