Suitability of various materials for porous filters in diffusion experiments

David Aldaba, Martin Glaus, Olivier Leupin, Luc Van Loon, Miquel Vidal, and Anna Rigol*

Abstract: The suitability of different porous materials (stainless steel, VYCOR® glass, Al₂O₃ and PEEK) for use as confining filters in diffusion experiments was evaluated by measuring the effective diffusion coefficients \( D_e \) of neutral (HTO) and ionic solutes \( D_w \) identified by measuring the effective diffusion coefficients \( D_e \) of neutral (HTO) and ionic solutes \( D_w \) evaluated in the materials in through-diffusion experiments. For stainless steel filters, the \( D_e \) values of the target solutes correlated satisfactorily with their bulk diffusion coefficient in water \( D_w \); thus, the diffusion process in the stainless steel filters was primarily controlled by the diffusivity of the solvated ions. For the remaining materials, the \( D_e \) and \( D_w \) values were also correlated for the target solutes, and the geometric factors were in the sequence: VYCOR® glass < Al₂O₃ < PEEK. Stainless steel and VYCOR® glass were the most appropriate materials because of their high \( D_e \) values, but a specific interaction of caesium with VYCOR® glass was hypothesised because the \( D_e \) values obtained for this solute were slightly higher than expected.

Keywords: Effective diffusion coefficient, porous filter, Archie’s relationship, geometric factor.

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1 Introduction

Based on international consensus [1] for the disposal of high-level radioactive waste (HLW) and spent fuel (SF), geological repositories have been promoted as the safest option to isolate the waste from the biosphere. In most versions of these repositories, the isolation of waste from the biosphere relies on passive multi-barrier systems. The space around the emplaced canisters is usually back-filled with swelling clay to reduce the hydraulic conductivity in the near field to values similar to those measured in the host rock [2, 3] and to ensure diffusion-controlled transport of radionuclides when the canisters are breached [4]. Thus, the development and application of laboratory methodologies for the characterisation of the diffusion of the radionuclides present in spent nuclear fuel and high-level waste in the compacted materials used in engineered barriers has become an important issue for long-term safety considerations.

The diffusive transport of radionuclides through a porous medium is quantified by the effective diffusion coefficient \( D_e \), which includes the effect of the geometric properties of the pores of the material on the diffusion of the target solutes. Therefore, \( D_e \) depends, among other terms, on the porosity accessible for diffusion, the pore diameter and the connectivity of the porous network [5]. In this sense, the diffusion of various radionuclides through compacted swelling clays and argillaceous rocks has been extensively evaluated in laboratory tests by obtaining the \( D_e \), primarily by through-diffusion experiments. The literature on through-diffusion experiments is vast but important references are: Molera and Eriksen [6], in which the diffusion of \( ^{22}\text{Na}^+ \), \( ^{85}\text{Sr}^{2+} \), \( ^{134}\text{Cs}^- \) and \( ^{57}\text{Co}^{2+} \) in compacted bentonite was studied as a function of different compaction densities; Van Loon et al. [7], who applied through-diffusion and the complementary out-diffusion method to study the diffusion of \( ^{36}\text{Cl}^- \) and \( ^{125}\text{I}^- \) in Opalinus Clay under different confining pressures; and Glaus et al. [8], who investigated the diffusion of HTO, \( ^{22}\text{Na}^+ \) and \( ^{36}\text{Cl}^- \) in compacted kaolinite, illite and montmorillonite as a function of the concentration of background electrolyte in the solution in contact with the clay sample.

The through-diffusion method usually requires a sandwiched filter-clay-filter experimental setup. The filters allow the tracer solution to enter and leave the clay samples [9] and maintain sample integrity. Moreover, in experiments with swelling clays (mainly compacted bentonites), stainless steel filters are often preferred because they can resist the high swelling pressures [6, 9]. The effect of the filters on the diffusion process is usually neglected; the prevailing argument for this neglect is that...
the solute diffusivity is much higher in filters than in clays. However, only a few studies have systematically evaluated radionuclide diffusion in stainless steel porous filters as a preliminary step [6, 9]. Thus, Molera and Eriksen [6] measured the diffusivities of solutes through stainless steel porous filters, and included these diffusivities in a model for solute diffusion in compacted bentonite. Gaul et al. [9] measured the $D_e$ values of various solutes in stainless steel porous filter discs before and after the use of these discs in diffusion experiments with clay minerals, and they concluded that used filters might impede solute diffusion because of the clogging of filter pores by clay. However, other factors, such as pore geometry or possible solute-filter interactions in unused filters, might also affect the $D_e$ values obtained in through-diffusion experiments and have not yet been evaluated. This evaluation could be performed by examining the diffusivities of a series of uncharged, positively and negatively charged solutes in a set of presumably inert porous media; this approach has not yet been fully and systematically tested in the literature.

Thus, in this work, the effect of the pore geometry (porosity, pore diameter and connectivity) on the quantification of $D_e$ by through diffusion [9], as well as the potential existence of solute-filter interactions, was evaluated in various materials for a series of solutes (HTO, Na$^+$, Cs$^+$, Sr$^{2+}$, Cl$^-$, SeO$_3^{2-}$), either radionuclides or, for safety, stable elements representative of relevant radionuclides. Archie’s relationship [10] was assessed for stainless steel porous filters with different pore diameters and porosities. In addition, the applicability of other materials (VYCOR® glass, Al$_2$O$_3$ and PEEK) for use as filters in through-diffusion experiments was also evaluated based on the relationship between the $D_e$ values obtained for the target solutes in these materials and the corresponding diffusion coefficients of the solutes in free water ($D_w$).

2 Experimental

2.1 Materials and reagents

The stainless steel filters used in this study were purchased from MOTT industrial division (Code 316L, Farmington, Connecticut, USA). The pore diameters of the filters were 10, 5, 2, 1 and 0.5 μm. The diameter of the discs was 25.4 mm, and the thickness was 1.5 mm. VYCOR® glass filter discs, composed of 96% SiO$_2$, 3% B$_2$O$_3$ and 0.4% Na$_2$O, were provided by Corning Incorporated (New York, USA). Typical pore sizes are of the order of 4 nm. Filters made from porous Al$_2$O$_3$ were provided by Metoxit (Thayingen, Switzerland). Only the particle size for the sintering process is known for this material (order of 5 μm); typical pore sizes are thus in the low μm range. The PEEK (Polyether ether ketone) filters were provided by Porogen (Woburn, MA, USA). The diameter of the disks was 25.6 mm in all cases and typical pore sizes for this material are in the 1 μm range. The thickness of the filter discs ranged from 1.00 to 1.03 mm for VYCOR® glass, from 1.50 to 1.53 mm for Al$_2$O$_3$, and from 1.65 to 1.68 mm for PEEK.

The porosities ($\varepsilon$) of all the filters tested were calculated from the weights of the dry and water-saturated filters.

P.A. grade CsCl, SrCl$_2$, and KCl were purchased from Fluka (Buchs, Switzerland). Radioisotopically pure solutions of HTO, $^{22}\text{Na}^+$, $^{36}\text{Cl}^-$, $^{134}\text{Cs}^+$ and $^{75}\text{SeO}_3^{2-}$ were obtained from Isotope Products Europe (Blaesg, Germany). Selenite was oxidised to selenate with NaClO based on similar procedures described in Gaul et al. [11] for the oxidation of iodide to iodate. The subsequent isolation of the reaction product by semi-preparative high-performance anion chromatography was performed with an IonPac AS18 column ( Dionex, Switzerland).

2.2 Through-diffusion experiments

For the through-diffusion experiments, the manufactured acrylic diffusion cells developed in a previous work [9] were used. Figure 1 shows a schematic of this experimental setup, which is composed of two solution reservoirs of 125 mL separated by the filter. Advection solute transport was prevented by the use of FKM fluoroelastomer flat seals (Angst + Pfister, Switzerland) that encased the filters. Once assembled, the cells were filled with approximately 115 mL of electrolyte solution (NaClO, 0.1 mol L$^{-1}$) and subjected to vacuum in a desiccator for approximately 15 min to evacuate any air trapped in the pore space of the filters. Then, the cells were allowed to stand for 24 h to ensure that the level of the solution was identical in both reservoirs. The solutions were stirred magnetically to maintain homogeneity. No special precaution was taken to fix the pH in the solutions. It can be assumed that pH was largely controlled by uptake of CO$_2$.

At the beginning of the experiments, 500 μL of a solution containing the target solute was added to one of the reservoirs (the source reservoir), and a similar amount of solution without the solute was simultaneously added to the other reservoir (the target reservoir). The initial concentration of solute added to the source reservoir was determined to facilitate quantification of the solute. This initial concentration was 1 MBq L$^{-1}$ for the radionuclides...
The concentrations of stable Cs$^+$ and Sr$^{2+}$ were measured by high-performance cation exchange chromatography (HPCEC) by a Dionex DX-600 system (Dionex, Switzerland) consisting of a metal-free GP50-2 quaternary gradient pump, a CD25A conductivity detector and an AS50 autosampler, equipped with a 9750 motor-driven Rheodyne injection valve and a 50 μl PEEK injection loop. The separation column was a 4 mm × 250 mm IonPac CS12A used in combination with a 4 mm × 50 mm IonPac CG12A guard column (Dionex, Switzerland). The eluent was 18 mmol L$^{-1}$ methanesulphonic acid at a flow rate of 1 mL min$^{-1}$. For stable Cl$^-$, a 4 mm × 250 mm IonPac AS16 separation column and a 4 mm × 50 mm IonPac AG16 guard column (Dionex, Switzerland) were used for the separation. The eluent was 0.02 mol L$^{-1}$ NaOH at a flow rate of 1 mL min$^{-1}$. Conductivity after electrochemical eluent suppression (autosuppression mode) was applied in both cases for analyte detection.

2.3.2 Experiments with VYCOR® glass, Al$_2$O$_3$ and PEEK filters

The concentration activities of HTO and $^{36}$Cl$^-$ present in the aliquots sampled were determined simultaneously by liquid scintillation with a Tri-carb 2250 CA counter (Canberra-Packard). The 250 μL sample aliquots were mixed with 10 mL of deionized water and 10 mL of scintillation cocktail (Ultima Gold XR, Canberra-Packard) for the measurements. $\beta$-$\beta$ discrimination was used in two energy windows: 0–9 keV for HTO and 9 to 710 keV for $^{36}$Cl$^-$. The lower energy window covered scintillation signals caused by the two radionuclides, while the upper window was specific for $^{36}$Cl$^-$. The activity of $^{36}$Cl$^-$ was thus determined unequivocally from the upper window, and this information was subsequently used for determining the activity of HTO.

2.3.1 Experiments with stainless steel filters

The concentrations of HTO and $^{22}$Na$^+$ present in the sampled aliquots were simultaneously determined by liquid scintillation with a Tri-carb 2250 CA counter (Canberra-Packard). The 250 μL sample aliquots were mixed with 10 mL of deionized water and 10 mL of scintillation cocktail (Ultima Gold XR, Canberra-Packard) for the measurements. $\beta$-$\beta$ discrimination was used in two energy windows: 0–9 keV for HTO and 9–160 keV for $^{22}$Na$^+$. The lower energy window covered scintillation signals caused by the two radionuclides, while the upper window was specific for $^{22}$Na$^+$. The activity of $^{22}$Na$^+$ was thus determined unequivocally from the upper window, and this information was subsequently used for determining the activity of HTO.
treated as being pseudo-linear because the concentration in the source reservoir did not change significantly during this time span. Subsequently, the curve evolved nonlinearly until the concentration of the solute was equal in both reservoirs. For example, Figure 2a shows the breakthrough curve of HTO for the experiments with VYCOR® glass filter, and Figure 2b shows the linear fit of the pseudo-linear phase.

The $D_e$ value for a given experiment was calculated by applying the following equation [9]:

$$D_e = \frac{a}{S C_0}$$

(1)

where $a$ (kBq s$^{-1}$ or mol s$^{-1}$) is the slope of the breakthrough curve obtained from a linear regression of the pseudo-linear phase, $L_e$ (m) and $S$ (m$^2$) are, respectively, the thickness and the cross-sectional area of the filter disc, and $C_0$ is the initial concentration (kBq m$^{-3}$ or mol m$^{-3}$) in the source reservoir.

No anomalies were detected in the diffusion experiments because the initial concentration of each given solute matched well (±5%) the sum of the final concentrations of the solute in the source and target reservoirs at the end of the experiment.

The global errors in the $D_e$ values were calculated by propagation of the errors associated with the parameters involved in Eq. (1). The errors were lower than 10% in all cases, and the main source of error originated from the calculation of the slope.

3 Results and discussion

3.1 Effective diffusion coefficients for the stainless steel porous filters

Table 1 summarises the effective diffusion coefficients of the target solutes for each pore diameter and porosity tested for the stainless steel porous filters.

For a given pore diameter and porosity, the results followed the sequence $D_e$(HTO) > $D_e$(Cs$^+$) ~ $D_e$(Cl$^-$) > $D_e$(Na$^+$) > $D_e$(Sr$^{2+}$); this sequence matches the sequence of the bulk diffusion coefficients in water ($D_w$) reported in the literature [12–14]. As shown in Figure 3a, the $D_e$ values obtained for all the examined solutes showed a satisfactory linear correlation ($0.92 < R^2 < 0.98$) with the respective $D_w$ values. The linear relation between $D_w$ and $D_e$ values was evaluated through the well-established formalism for the diffusion of a solute in a saturated porous system [15]:

$$D_e = \frac{\varepsilon}{G}D_w$$

(2)

where $G$ is the overall geometric factor, which accounts for the geometric properties of the pore network, such as the constrictivity and tortuosity. $G$ values were calculated

![Fig. 2: (a) Breakthrough curve for HTO and (b) pseudo-linear phase of the breakthrough curve in the target reservoir for the experiments with the VYCOR® glass filter.](image-url)
In addition to the porosity and the size distribution of pores, the topology of the pore space is a crucial property for flow and transport in porous media. The way that pores are interconnected (pore connectivity) may be even more significant than the number and size of the pores. As shown in Table 1, the porosity increased with pore size in the stainless steel filters up to the 5-μm filters, but this increase was not observed for the 10-μm filters, which had a lower porosity. The opposite trend was observed for the G values. This fact might be related to the production process for these filters, in which a higher pore diameter did not consistently lead to a higher porosity and connectivity. The $D_e$ of a given solute increased with the filter porosity (up to the maximum $D_e$ values for porosities of approximately 0.30-0.32) but not with the pore size. Therefore, the experimental results indicated that the filter pore size is not the critical parameter governing the $D_e$ values. Instead, the dependence between $D_e$ and the porosity values can be examined based on Archie’s empirical relationship in its general form [10, 18]:

$$D_e = A \cdot \varepsilon^m$$  \hspace{1cm} (4)

where $A$ is a pre-exponential factor, which is expected to equal $D_w$ for an inert porous medium in the limiting case of $\varepsilon \to 1$; $m$ is the empirical cementation exponent, which with the porosity, is a measure of how the pore space is arranged in terms of pore size and connectivity (pore geometry) [10]. Thus, the cementation exponent characterises the tendency of the pore network to increase the diffusive resistivity. Note the formal analogy between Eq. (2) and Eq. (4) for the identity of $A$ and $D_w$, from which it follows that $G = \varepsilon^{1-m}$. Archie’s relation can also be applied to simultaneously derive the $m$ and $A$ values for each solute through the log transformation of Eq. (4),

$$\log D_e = \log A + m \cdot \log \varepsilon$$  \hspace{1cm} (5)

thus, $\log D_e$ can be correlated to $\log \varepsilon$ for every solute. The $A$ and $m$ values can then be derived from the intercept and slope of the regression curve, respectively. As shown in Figure 3b, all the correlations were excellent ($R^2 > 0.93$), and Table 2 summarises the $A$ and $m$ values calculated for all the solutes. The corresponding $D_w$ values from the literature are also included for comparison.

Reasonably constant values of $m$ were obtained for all the solutes, from 1.7 to 1.9; this result suggests that the pore geometry is quite homogeneous in the stainless steel filters for the various pore sizes and porosities evaluated.

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**Fig. 3:** Archie’s correlations in the stainless steel porous filters: (a) $D_e$ as a function of $D_w$ and (b) log $D_e$ as a function of log $\varepsilon$. The symbols correspond to the experimental data, and the lines correspond to the fits.
Table 2: The \(m\) factor and \(D_w\) values derived from the correlations of 
\(\log D_e\) as a function of \(\log \varepsilon\) and the literature \(D_w\) values for each 
solute. The standard errors for \(m\) and \(D_w\) are given within the 
parentheses.

<table>
<thead>
<tr>
<th>Solute</th>
<th>Derived</th>
<th>Literature (D_w)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(m) factor</td>
<td>(A) ((10^{-10} \text{ m}^2 \text{ s}^{-1}))</td>
</tr>
<tr>
<td>HTO</td>
<td>1.7 ((0.2))</td>
<td>19 ((5))</td>
</tr>
<tr>
<td>Cs(^{+})</td>
<td>1.8 ((0.2))</td>
<td>18 ((5))</td>
</tr>
<tr>
<td>Cl(^{-})</td>
<td>1.8 ((0.2))</td>
<td>19 ((6))</td>
</tr>
<tr>
<td>Na(^{+})</td>
<td>1.8 ((0.2))</td>
<td>13 ((3))</td>
</tr>
<tr>
<td>Sr(^{2+})</td>
<td>1.9 ((0.2))</td>
<td>8 ((2))</td>
</tr>
</tbody>
</table>

\(^{a}\) \[14], \(^{b}\) \[15], \(^{c}\) \[13].

Furthermore, the \(m\) values obtained in these filters were 
lower than the values reported in the literature for natural 
samples and indicated a less complex pore geometry 
because of a more ordered aggregation of particles. Values 
of \(m\) in the literature ranged from theoretical values 
of 1.35, for a homogeneously connected isotropic medium 
of spherical particles \[19\], to higher values (up to 3), 
corresponding to the more complex pore geometries present 
in natural samples. For example, a \(m\) value of 2.3 was 
estimated for limestone and sandstone \[19\], \(m\) = 2.5 was 
estimated for Opalinus clay and Boom clay \[7\] and \(m\) = 3 
was estimated for Oxfordian limestones and argillite \[20\].

The \(A\) values for Cs\(^{+}\), Cl\(^{-}\), Na\(^{+}\) and Sr\(^{2+}\) deduced 
from the respective correlations were similar to the 
literature values of \(D_w\): thus, these values validated the assumption 
of bulk water properties for the pore fluid, and suggested 
that the stainless steel filters behaved as inert porous media 
and did not interact with the solute. This result is not 
given for reactive porous media, such as clays, in which \(A\) 
does not equal \(D_w\), as in montmorillonite (see, e.g., Glaus 
et al., 2010). Therefore, in view of the obtained results, 
unused stainless steel filters are considered to be appropriate 
for use as confining porous filters in through-diffusion 
experiments. Additionally, Archie’s relationship might also 
be used to roughly estimate the \(D_w\) values of a given solute 
in stainless steel porous filters.

3.2 Effective diffusion coefficients 
for VYCOR\(^{\circledR}\) glass, \(\text{Al}_2\text{O}_3\) and PEEK porous filters

Table 3 shows the measured effective diffusion coefficients 
of HTO, Cs\(^{+}\), Cl\(^{-}\) and SeO\(_4^{2-}\). In contrast to the 
experiments with steel filters, the \(D_e\) values for VYCOR\(^{\circledR}\) glass, 
\(\text{Al}_2\text{O}_3\) and PEEK porous filters were measured only for the 
single porosity given in Table 3. In PEEK filters, the 
sequence of \(D_e\) values was \(D_e(\text{HTO}) > D_e(\text{Cl}^-) \approx D_e(\text{Cs}^+) > D_e(\text{SeO}_4^{2-})\), which can be explained roughly by the 
diffusivity of the solvated ions, as already observed in stain-
less steel filters. In contrast, the sequence of \(D_e\) values 
observed in the experiments with the VYCOR\(^{\circledR}\) glass 
and \(\text{Al}_2\text{O}_3\) filters did not follow the same trend because the \(D_e\) 
value for Cs\(^{+}\) was slightly higher than that expected from 
the hydrated radius of Cs\(^{+}\). Gimmi and Kosakowski \[21\] 
also observed that some sorbing cations, such as caesium, 
diffused at larger rates than expected in clay materials; 
this faster diffusion was attributed to surface diffusion, 
a process that occurs in parallel to pore diffusion. This 
interaction process could also be hypothesised to occur 
with caesium in \(\text{Al}_2\text{O}_3\) and especially VYCOR\(^{\circledR}\) glass 
filters with the smallest pores; however, no evidence of this 
interaction could be deduced from the mass balance of the 
caesium concentration in the experiments performed with 
these two materials. Additionally, note that the pore sizes 
in these two materials are of completely different orders of 
magnitude, so completely different preconditions exist for 
such a process to be relevant.

Table 3: The porosities (\(\varepsilon\)), effective diffusion coefficients and 
geometric factors (\(G\)) obtained through-diffusion experiments 
with VYCOR\(^{\circledR}\) glass, \(\text{Al}_2\text{O}_3\) and PEEK porous filters.

<table>
<thead>
<tr>
<th>Material</th>
<th>(\varepsilon)</th>
<th>(D_e(10^{-10} \text{ m}^2 \text{ s}^{-1}))</th>
<th>(G)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HTO</td>
<td>0.20</td>
<td>2.5</td>
<td>1.8</td>
</tr>
<tr>
<td>(\text{Al}_2\text{O}_3)</td>
<td>0.30</td>
<td>1.9</td>
<td>1.5</td>
</tr>
<tr>
<td>PEEK</td>
<td>0.36</td>
<td>1.4</td>
<td>1.0</td>
</tr>
</tbody>
</table>

\(\text{SeO}_4^{2-}\)

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Table 3 shows the measured effective diffusion coefficients 
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\(\text{SeO}_4^{2-}\)
Another important aspect is that the highest $D_e$ values were obtained for VYCOR® glass, which had the lowest porosity, whereas the lowest values were obtained for PEEK, which had the highest porosity. In contrast, as shown in Table 3, the highest values of $G$ were obtained for PEEK filters, with the highest porosity, whereas the lowest values were obtained for VYCOR® glass filters, with the lowest porosity. This result indicates that the $m$ values (see Archie’s relation) are not the same for these materials (or according to the equation, $G = \varepsilon^{1-m}$ and suggests that porosity alone is insufficient to explain the differences in solute diffusion in these materials, and the geometry of the pores, including the pore connectivity, might also have an effect.

As shown in Figure 4, despite these irregularities, a fairly linear relation was obtained for the three porous materials between the $D_e$ values of each solute and the respective $D_w$ values obtained from the literature [12–14], except for SeO$_4^{2-}$, for which a $D_w$ value of $7 \times 10^{-10}$ m$^2$ s$^{-1}$, obtained in this work by a capillary method [22], was used. The correlation in the PEEK filters was excellent ($R^2 = 0.95$), but was slightly worse in VYCOR® glass ($R^2 = 0.89$) and Al$_2$O$_3$ ($R^2 = 0.91$) filters because of the different pattern of the $D_e$ values obtained for Cs$^+$ relative to the remaining solutes. The correlations improved for the two materials ($R^2 = 0.97$ for VYCOR® glass and $R^2 = 0.99$ for Al$_2$O$_3$) if the Cs$^+$ values were excluded; this improvement indicated that interaction processes possibly affected the diffusion of Cs$^+$ in these materials. Nevertheless, these processes must be considered to be merely speculative because of the few number of points considered.

4 Conclusions

This work demonstrated that testing the correlation between $D_e$ and $D_w$ values in a series of uncharged, positively and negatively charged solutes in presumably inert porous media is a useful strategy for the evaluation of the suitability of a given material for use as a porous filter in through-diffusion experiments. This strategy allows information to be obtained about the potential interactions of the solutes with the filter and anticipates the possible effects of the pore geometry of the filter material on the $D_e$ values obtained for different solutes, as a preliminary step in the design of diffusion experiments. Thus, filter materials that result in good correlations and that allow a good estimate of the $D_w$ values for a solute from Archie’s relationship are suitable for diffusion experiments. However, there are also other criteria for the selection of filter materials, such as the cementation factor in Archie’s relationship, when dealing with different porosities of the same material, or the geometrical factor, which permits comparisons among materials. The lower the $D_e$ values obtained for a filter, the less suitable the filter material is. The mechanical stability or the presence of solute-material interaction processes are also other criteria to consider.

Therefore, for high $D_e$ values, stainless steel filters with a pore size of 2 to 5 μm or VYCOR® glass filters would be the most suitable materials. However, VYCOR® glass may have a potential interaction with Cs$^+$ whereas stainless steel filters showed a good correlation for a wide range of solutes.

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