BELBaR Project

STUDY OF ^{85}Sr TRANSPORT THROUGH A COLUMN FILLED WIH CRUSHED GRANITE IN PRESENCE OF BENTONITE COLLOIDS

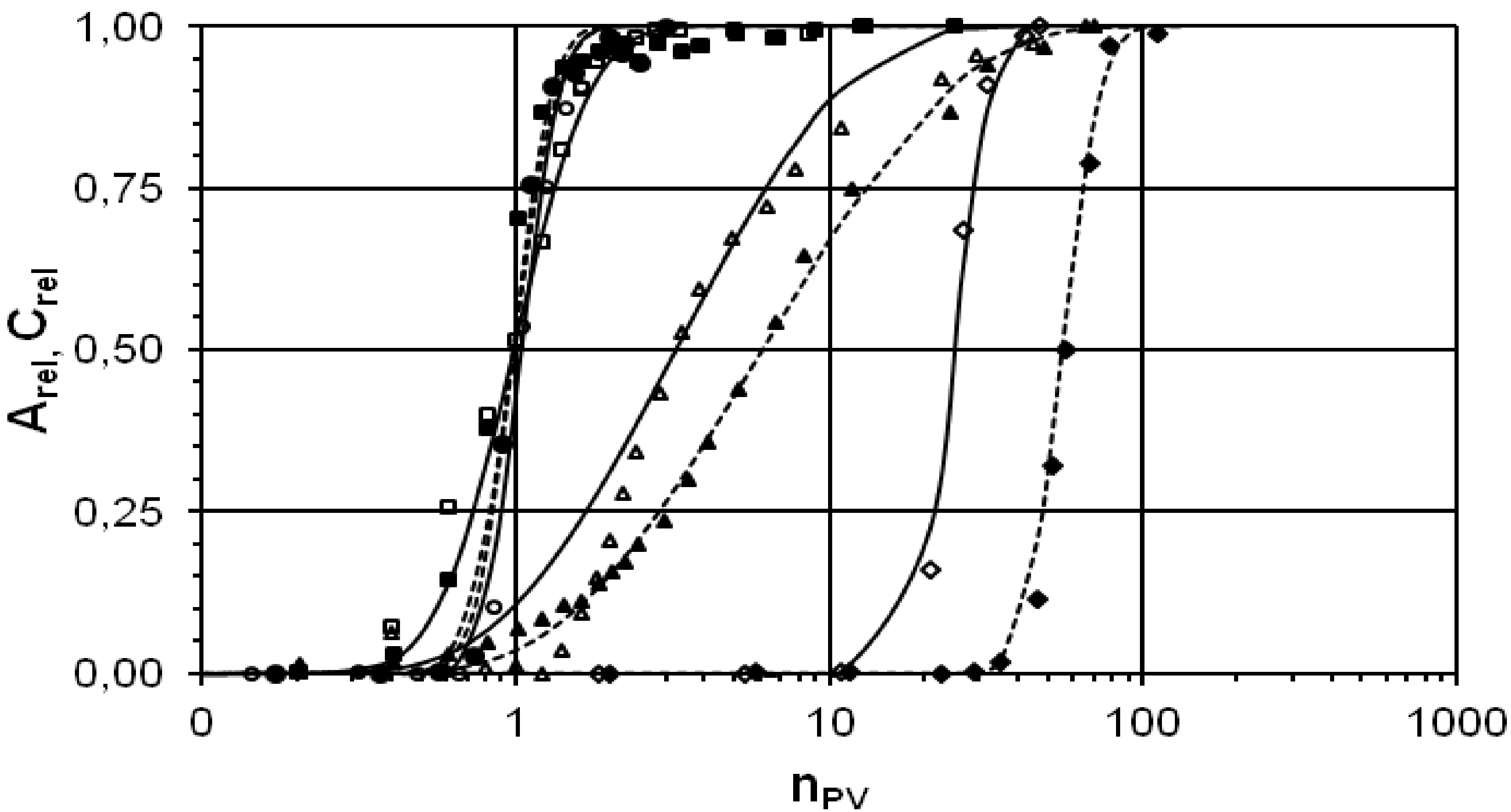
WP3

Kateřina Videnská, Radek Červinka

ÚJV Řež, a. s., 2014

Previous experiments

- Dynamic column experiments with different tracers without presence of colloids.



Previous experiments

- Dynamic column experiments with different tracers without presence of colloids.

What have we focused on?

- Column experiments with radiocolloids of ^{85}Sr.
- Hypotesis:
  - Colloid particles will pass through granite without retardation.
  - ^{85}Sr will pass through column together with colloids particles without retardation.
  - Part of ^{85}Sr will pass through column with colloids, part of ^{85}Sr will interact with granite.
  - ^{85}Sr transport in presence of colloids will be similar as ^{85}Sr transport without presence of colloids.

Goals of experiments

- Study of radiocolloids, strontium and colloids transport through column filled with crushed granite
- Influence of bentonite colloids on strontium migration through granite

EXPERIMENTAL

Solid and liquid phase

Melechov granite

- Mineralogical composition:
  - main: quartz, feldspar
  - minority: mica (muscovite), chlorite
- Depth: 97.5-98.7 m
- Fraction: 0.125-0.63 mm

Deionized water (DW)

- Previous column experiments without presence of colloids was performed in SGW ⇒ difficult to compare
- Synthetic granitic water (SGW) is not suitable for long-term stability of bentonite colloids.

Bentonite colloids suspension

- Na^+ form of non-activated purified Bentonite 75, denoted B75 deionized water
- c = 100 mg/l
- hydrodynamic size of bentonite particles: 500 nm (PCCS measurement)

Tracers

- HTO solution
- $SrCl_2$ solution
  - $c = 1 \cdot 10^{-6}$ mol/l
  - spiked by ^{85}Sr, activity 0.7 kBq/ml
- Bentonite colloid suspension
  - c = 100 mg/l
- Radiocolloid suspension
  - c ($SrCl_2$) = $1 \cdot 10^{-6}$ mol/l, spiked by ^{85}Sr, activity 0.7 kBq/ml
  - c (B75) = 100 mg/l
  - contact time between $^{85}SrCl_2$ and colloids to ensure equlibrium: 7 days
  - ratio between V (B75) and V($^{85}SrCl_2$) = 1:1
  - 90% of ^{85}Sr sorbed on colloids

Sorption experiments

$$K_D = \frac{(A_0 - A)}{A}\frac{V}{m}$$

- **Static batch method**
  - sorption distribution coefficient K_d (ml/g)
- **Tracer**
  - $^{85}SrCl_2$
- **Solid phase**
  - crushed Melechov granite, grain size 0.125-0.63 mm
  - mica mineral
- **Liquid phase**
  - deionized water
- **Time of contact phase**
  - 9 days
- **Aerobic conditions**
- **Phase ratio**
  - s : l = 1 : 20

Dynamic experiments

- **Column arrangement**
  - retardation coefficient *R*
  - sorption distribution coefficient K_d (ml/g)
- **Tracer**
  - HTO solution
  - $SrCl_2$
  - B75 colloids
  - radiocolloids (B75 + $^{85}SrCl_2$)
- **Solid phase**
  - crushed Melechov granite
  - grain size 0.125-0.63 mm
- **Liquid phase**
  - deionized water

- **Setup of column experiments**
  - plastic columns
  - 4-double head peristaltic pump
  - 8-outlet manifold
  - reservoir of liquid phase
  - continuous inlet of liquid phase

Dynamic experiments

Evaluation of column experiments

- **Breakthrough curve**
  - S-shape with inflection point (see index *i*)
  - x-axis: the number of pore volume (n_{PV})
  - y-axis: the strontium activity (A_{rel}) or colloid concentration (c_{rel}) measured at outlet of column

Modelling of breakthrough curve

- CXTFIT Code (STANMODE, v. 2.08)
- 1D transport model
- input transport parameters: dispersion coefficient *D*, velocity *v*, *R*
- estimated parameters: *D*, *v*

Retardation coefficient *R*

- $c_{rel,i} = 0.5$, $R = n_{PV,i}$

Distribution coefficient K_d

$$K_d = \frac{\theta}{\rho}(R-1)$$

where K_d is distribution coefficient (ml/g), θ is porosity, ρ is bulk density, *R* is retardation coefficient

RESULTS

Sorption experiments

- Static batch method

- First information about strontium sorption on granitic rock in deionized water whithout presence of colloids particles.

- Mineralogical analysis of granite:
    - quartz (32 %), orthoclase (31 %), plagioclase (18 %)
    - **mica (14 %), chlorite (5 %)**

- Assumption: High strontium sorption on mica minerals $\Rightarrow$ comparing the strontium sorption on granite and mica.

Sorption experiments

Influence of liquid phase composition on strontium sorption

- Sorption on granite: s : l = 1 : 20

  $c(SrCl_2) = 1 \cdot 10^{-6}$ mol/l, SGW

  K_d = 15.3 ml/g

- Sorption on granite: s : l = 1 : 20

  $c(SrCl_2) = 1 \cdot 10^{-6}$ mol/l, deionized water (DW)

  K_d = 30.6 ml/g

  - K_d (DW) = 30.6 ml/g vs K_d (SGW) = 15.3 ml/g
  - Higher K_d on granite in deionized water
  - **Competition effect of other ions in SGW leads to decrease of Sr retention.**

Sorption experiments

Influence of granite composition on strontium sorption

- Sorption on granite: s : l = 1 : 20, 14 % mica

  c($SrCl_2$) = 1·10^{-6} mol/l, deionized water

  K_d = 30.6 ml/g, sorption yield 60 %

- Sorption on mica: s : l = 1 : 20

  c($SrCl_2$) = 1·10^{-6} mol/l, deionized water

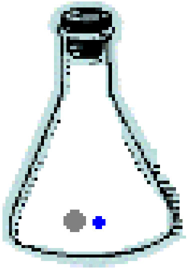
  K_d = 20.6 ml/g, sorption yield 50 %

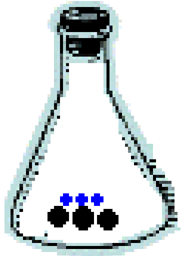
Mica   Granite

m = 0.1 g

Sorption yield: 50 %

$n(Sr)_{sorbed} = 1 \cdot 10^{-9}$ mol





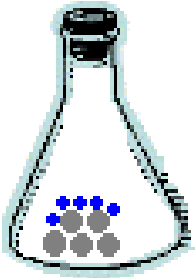
m = 0.5 g

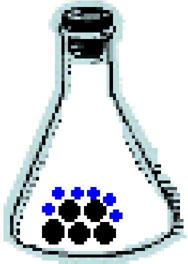
Sorption yield: 60 %

$n(Sr)_{sorbed} = 6 \cdot 10^{-9}$ mol

relative to 1 g of material

$n\ (Sr)_{sorbed} = 1 \cdot 10^{-8}$ mol



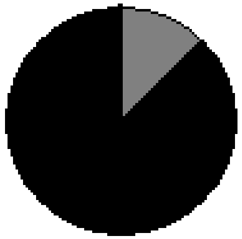


$n\ (Sr)_{sorbed} = 1.2 \cdot 10^{-8}$ mol

1 g of granite contains 0.14 g of mica

12 % of Sr was sorbed on mica in granite

88 % of Sr was sorbed on other minerals in granite

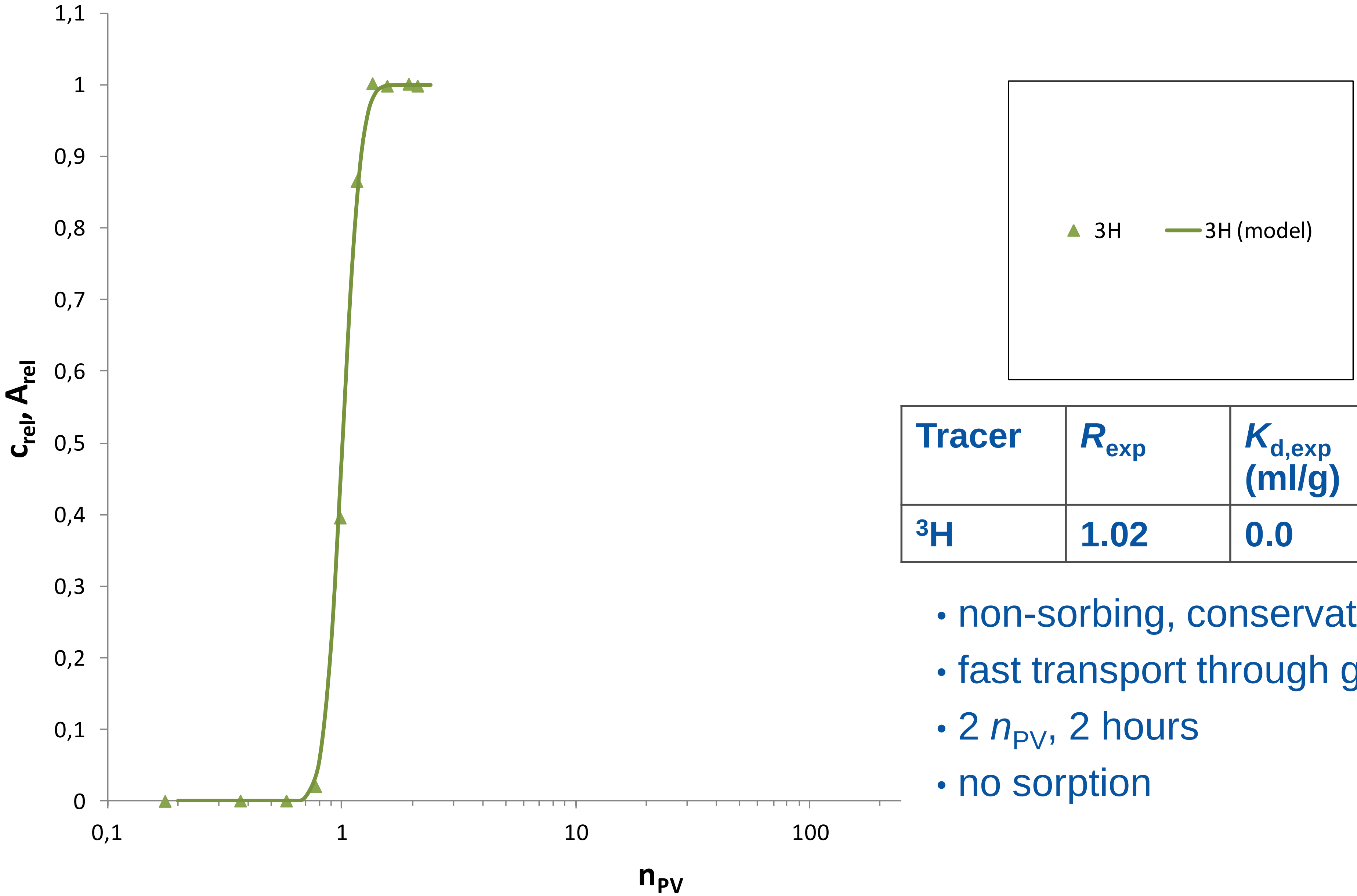


⇒ mica is not dominant sorbent of strontium in granitic rock

Dynamic experiments

- Tracers:
    - ^{3}H as HTO
    - bentonite colloids (B75)
    - $^{85}SrCl_2$
    - radiocolloids (B75+$^{85}SrCl_2$)

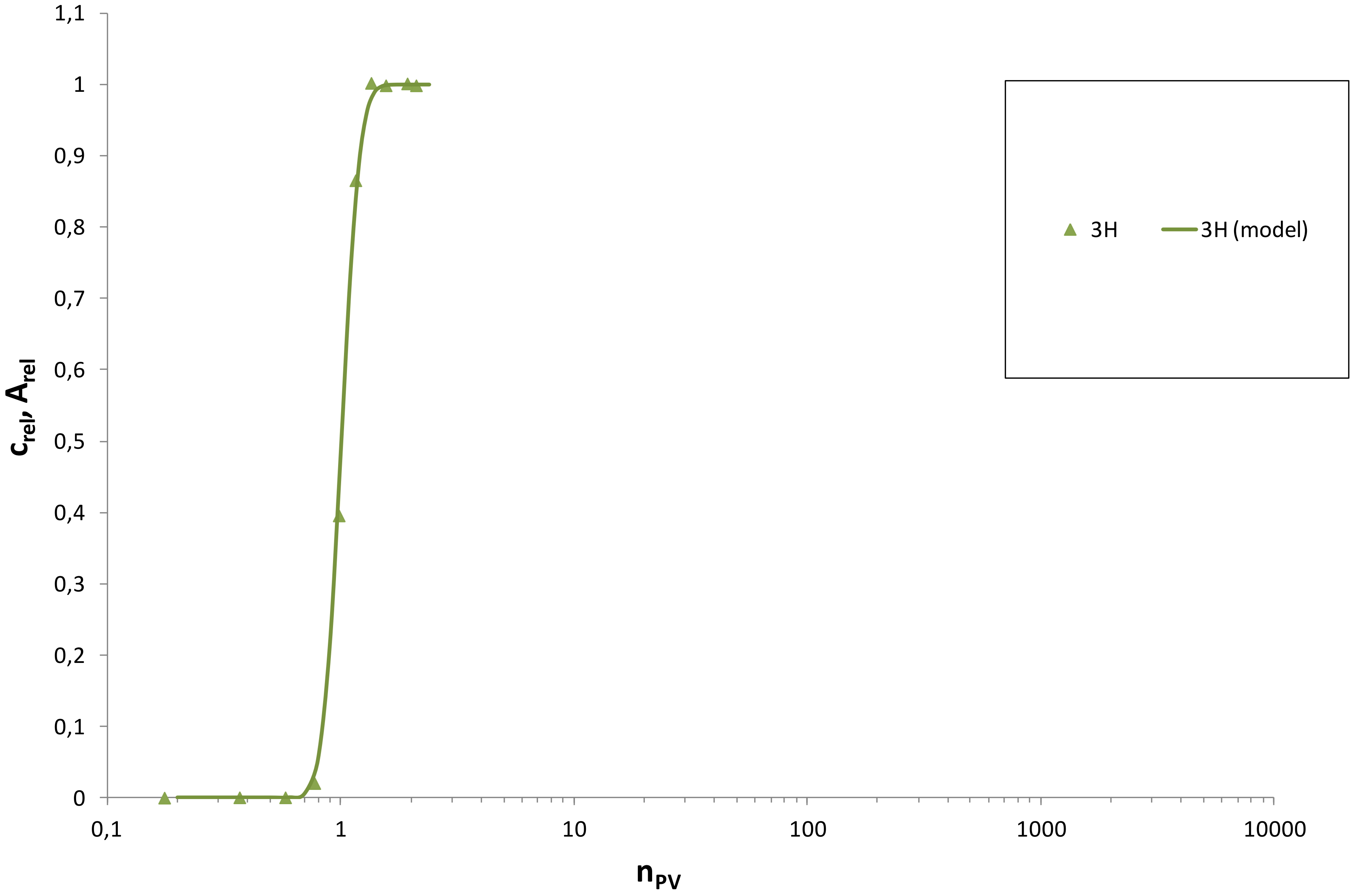
Dynamic experiments: ^3H



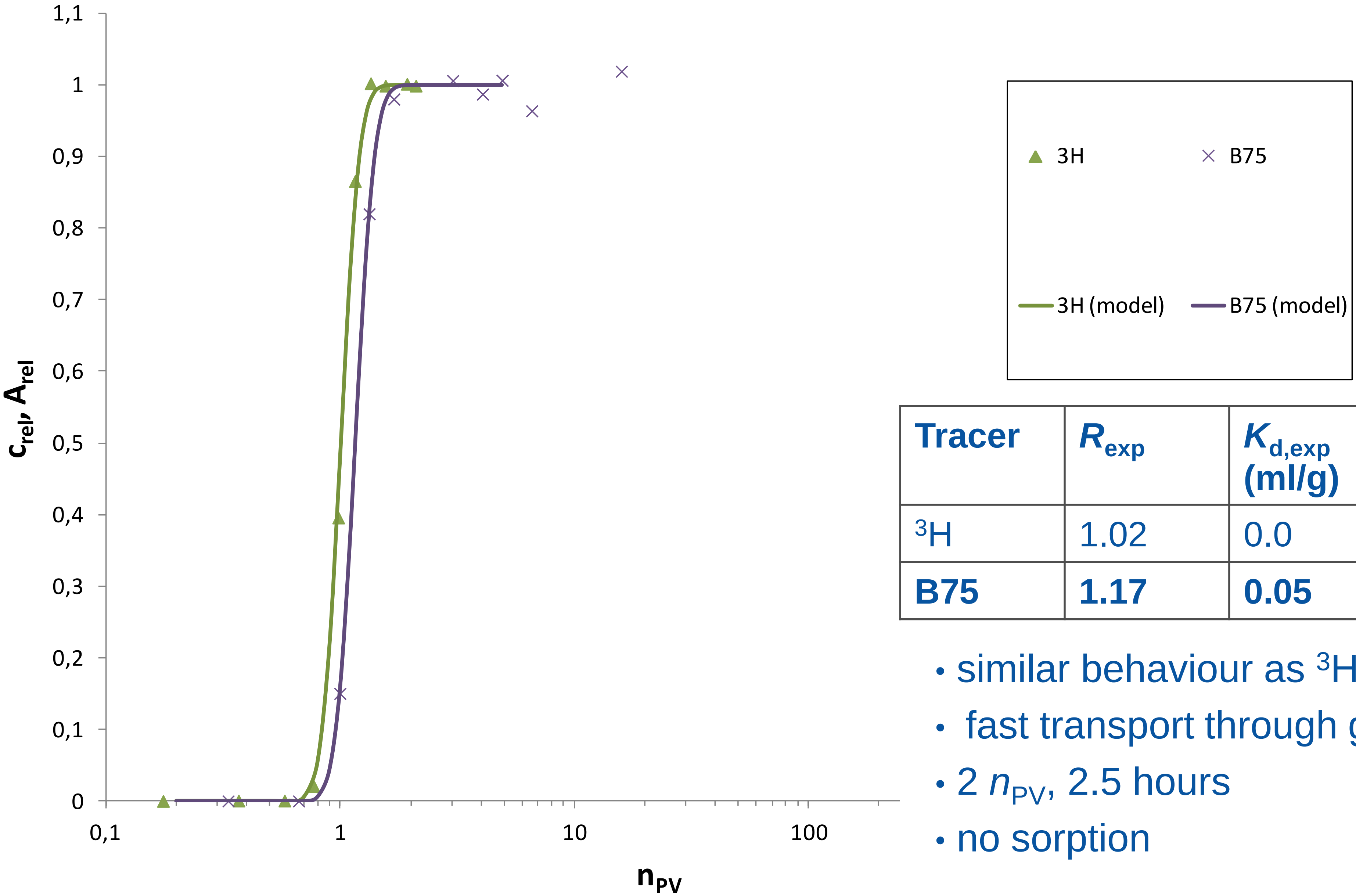
Tracer	R_{exp}	$K_{d,exp}$ (ml/g)	D_{model} (cm²/d)
^3H	1.02	0.0	0.0100

- non-sorbing, conservative tracer
- fast transport through granite
- 2 n_{PV}, 2 hours
- no sorption

Dynamic experiments: B75 colloids



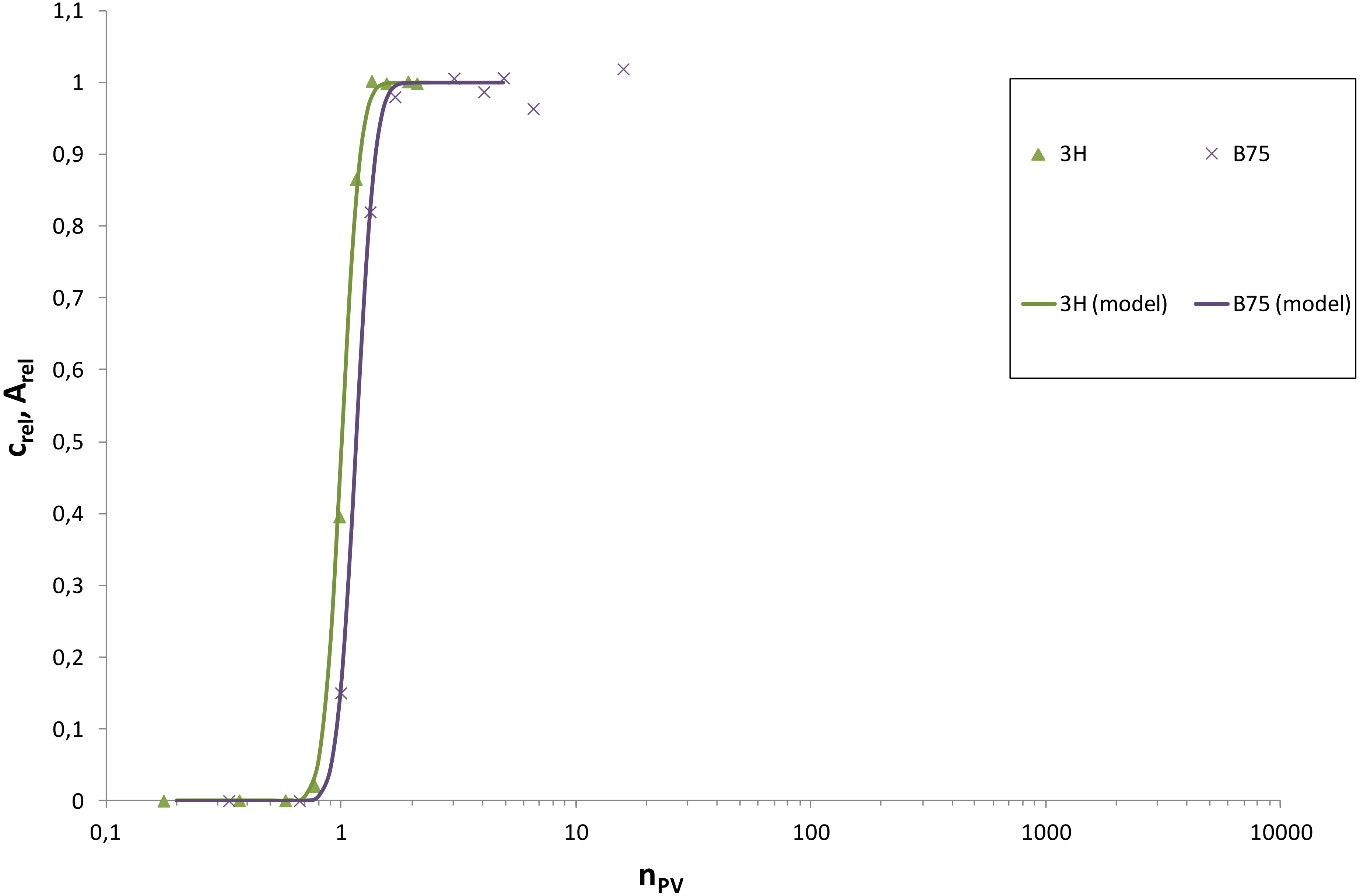
Dynamic experiments: B75 colloids



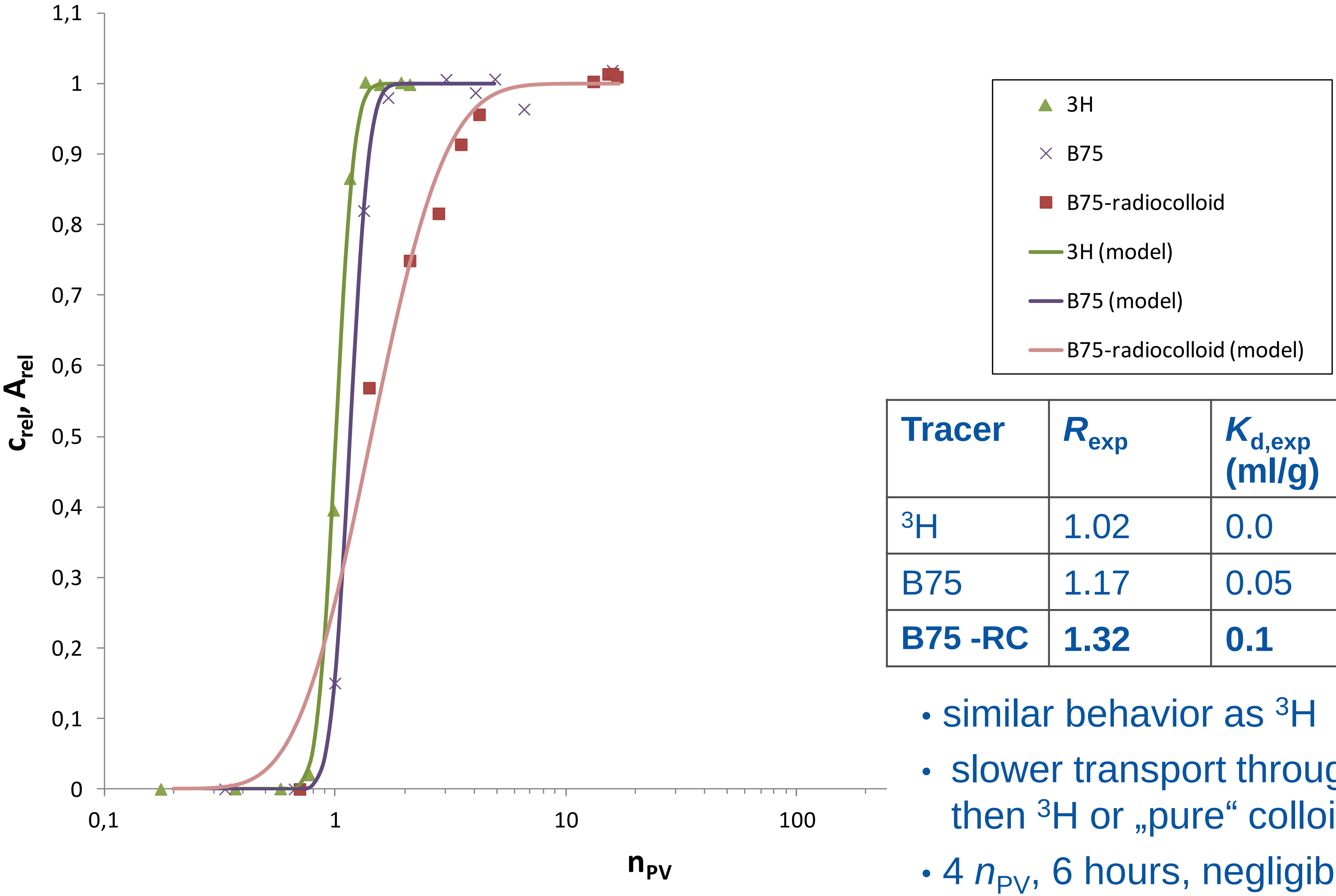
Tracer	R_{exp}	$K_{d,exp}$ (ml/g)	D_{model} (cm^2/d)
^3H	1.02	0.0	0.0100
B75	**1.17**	**0.05**	**0.0108**

- similar behaviour as ^3H
- fast transport through granite
- 2 n_{PV}, 2.5 hours
- no sorption

Dynamic experiments: B75 colloids (radiocolloids)



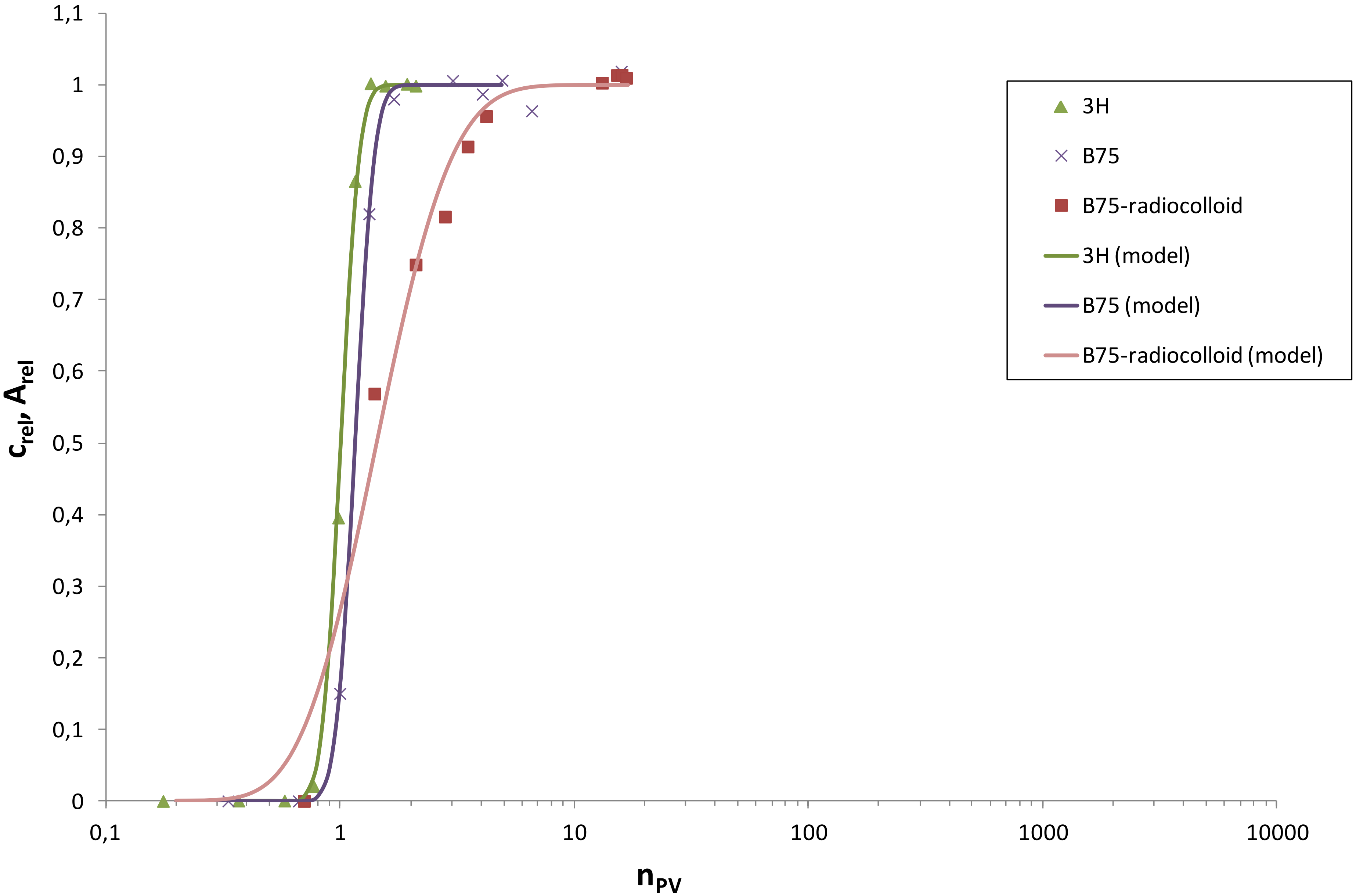
Dynamic experiments: B75 colloids (radiocolloids)



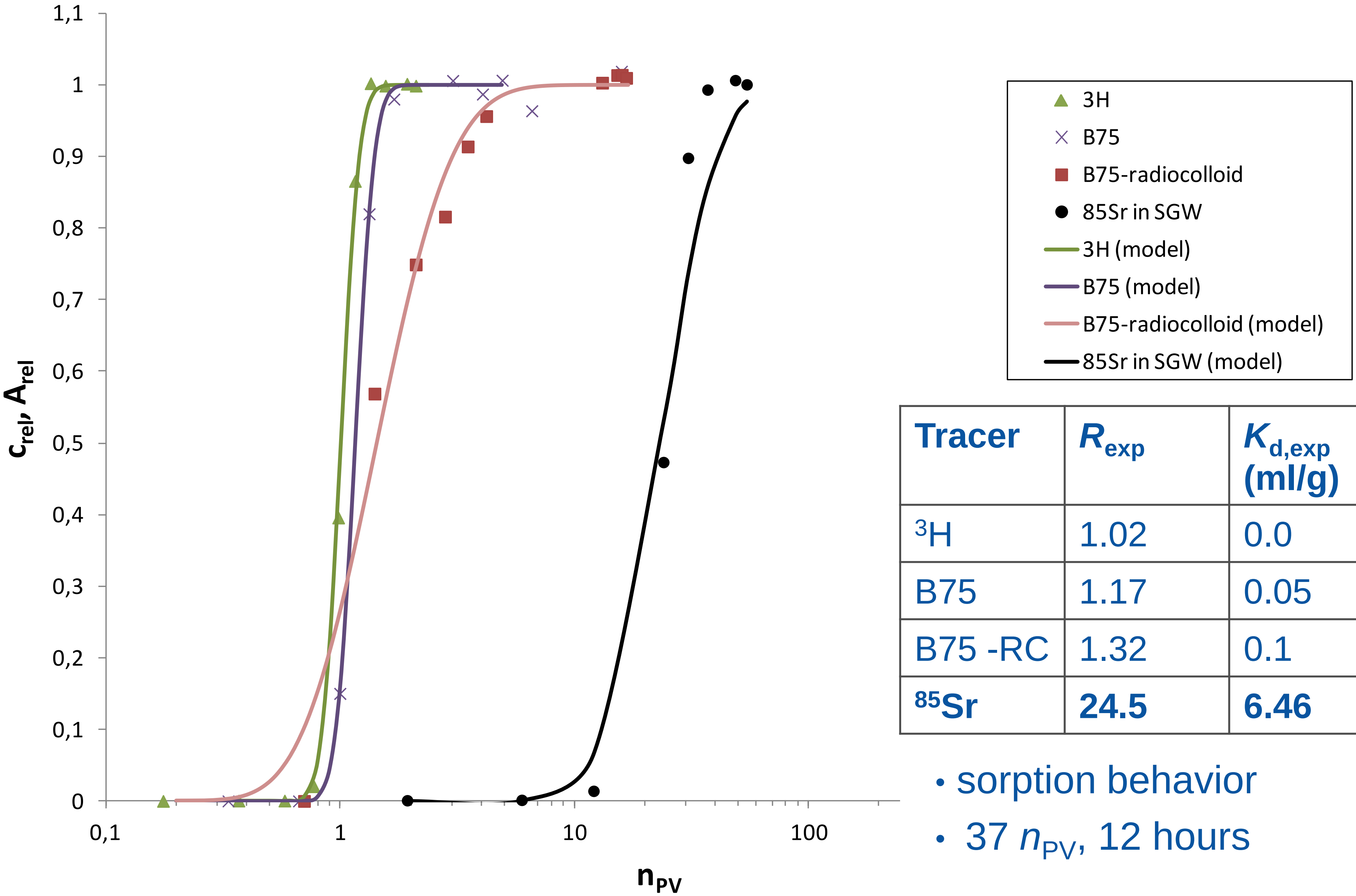
Tracer	R_{exp}	$K_{d,exp}$ (ml/g)	D_{model} (cm^2/d)
^3H	1.02	0.0	0.0100
B75	1.17	0.05	0.0108
B75 -RC	**1.32**	**0.1**	**0.1430**

- similar behavior as ^3H
- slower transport through granite then ^3H or „pure“ colloids
- 4 n_{PV}, 6 hours, negligible sorption

Dynamic experiments: ^{85}Sr (in SGW)



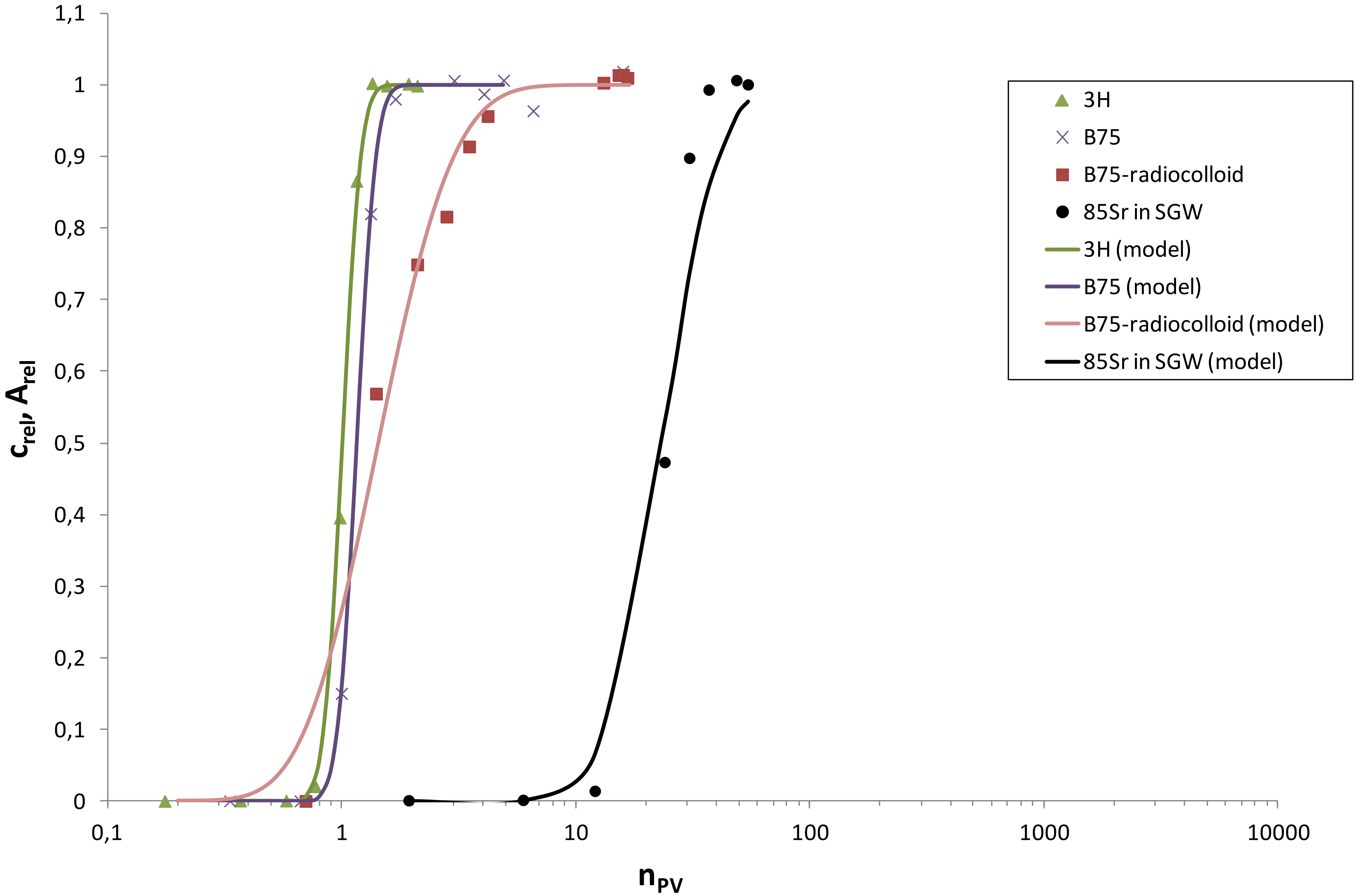
Dynamic experiments: ^{85}Sr (in SGW)



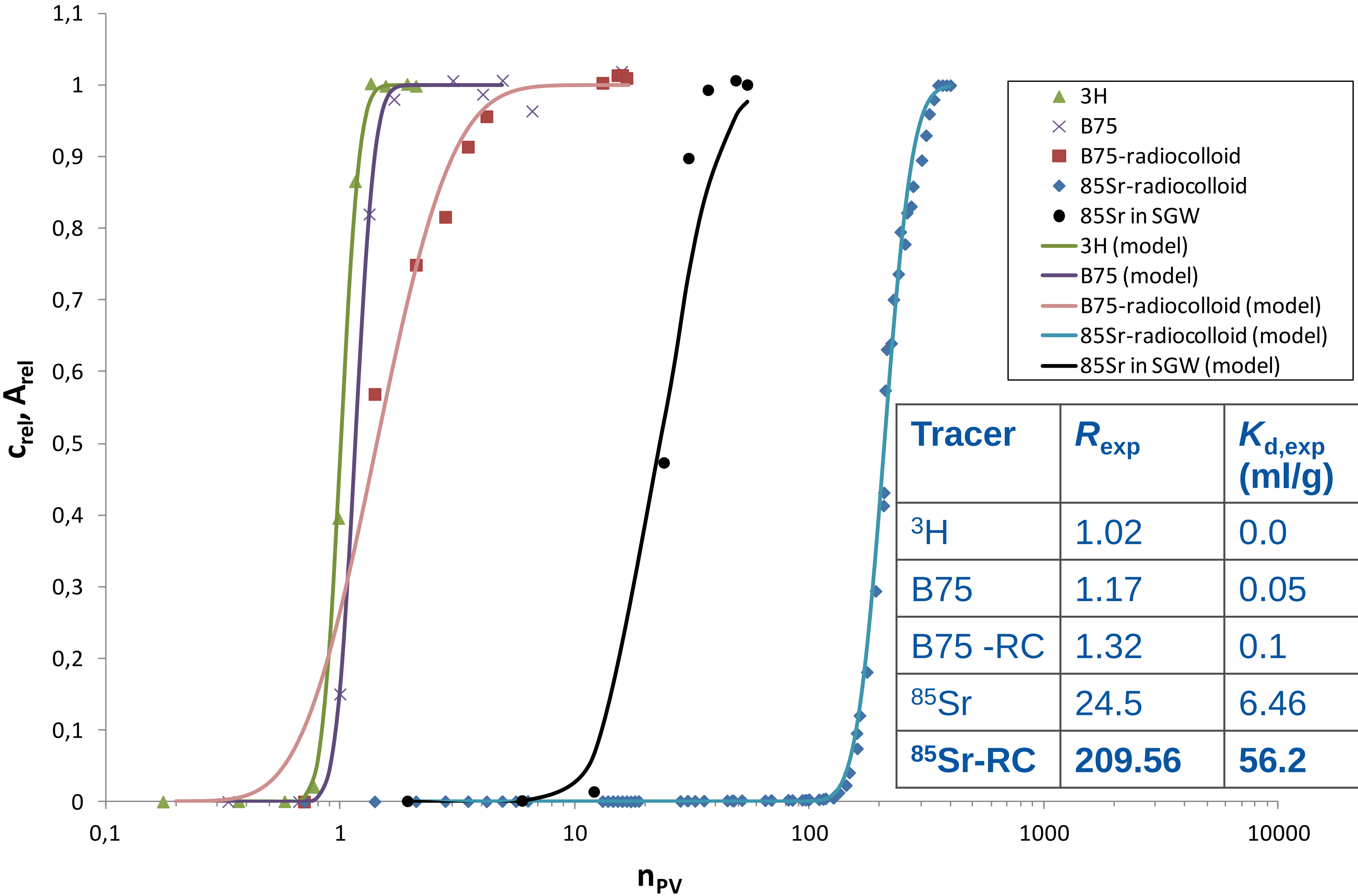
Tracer	R_{exp}	$K_{d,exp}$ (ml/g)	D_{model} (cm²/d)
^{3}H	1.02	0.0	0.0071
B75	1.17	0.05	0.0108
B75 -RC	1.32	0.1	0.1430
^{85}Sr	**24.5**	**6.46**	

- sorption behavior
- 37 n_{PV}, 12 hours

Dynamic experiments: ^{85}Sr (radiocolloids)

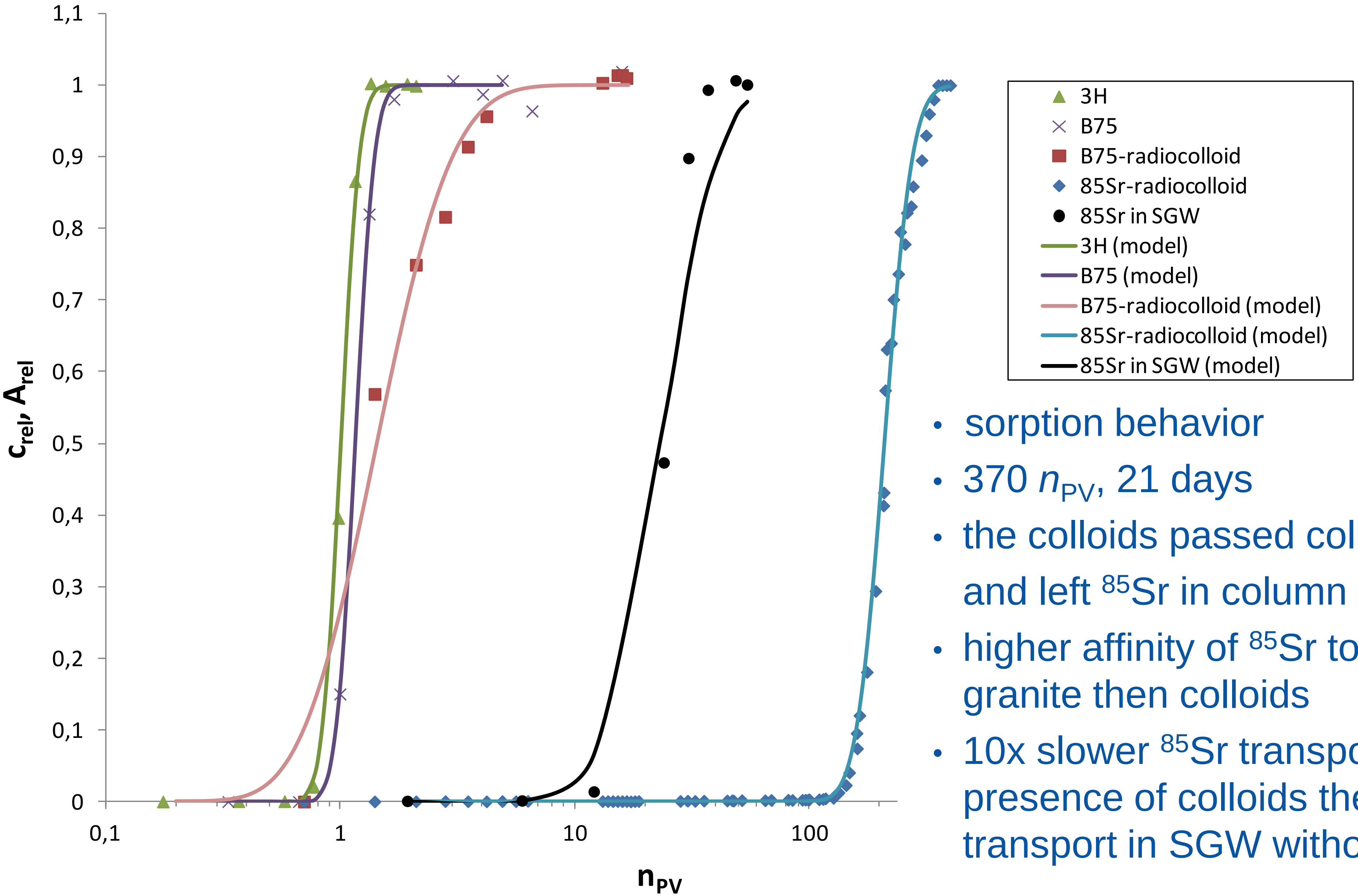


Dynamic experiments: ^{85}Sr (radiocolloids)



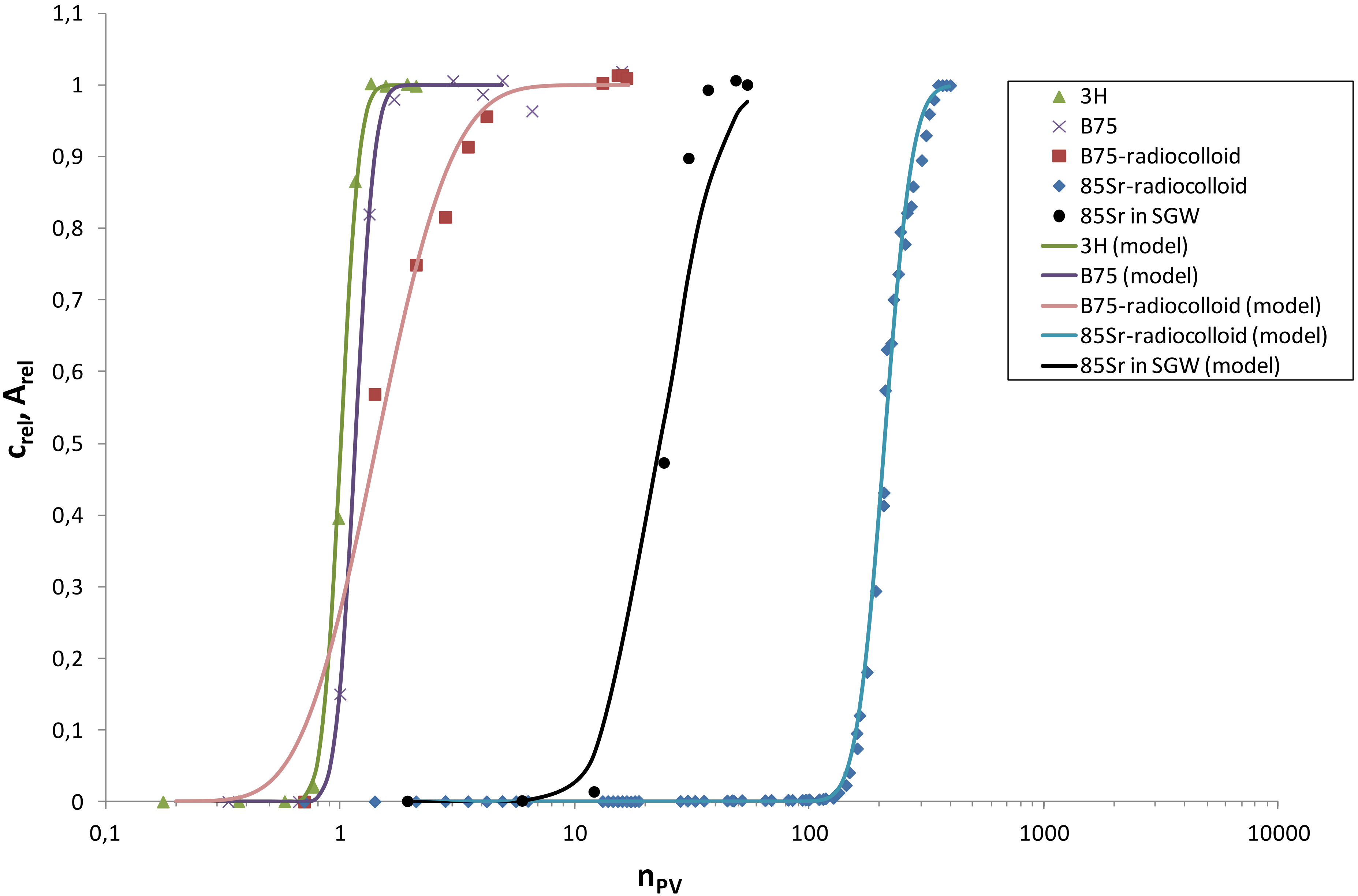
Tracer	R_{exp}	$K_{d,exp}$ (ml/g)	D_{model} (cm^2/d)
^{3}H	1.02	0.0	0.0071
B75	1.17	0.05	0.0108
B75 -RC	1.32	0.1	0.1430
^{85}Sr	24.5	6.46	
^{85}Sr-RC	**209.56**	**56.2**	**0.0231**

Dynamic experiments: ^{85}Sr (radiocolloids)

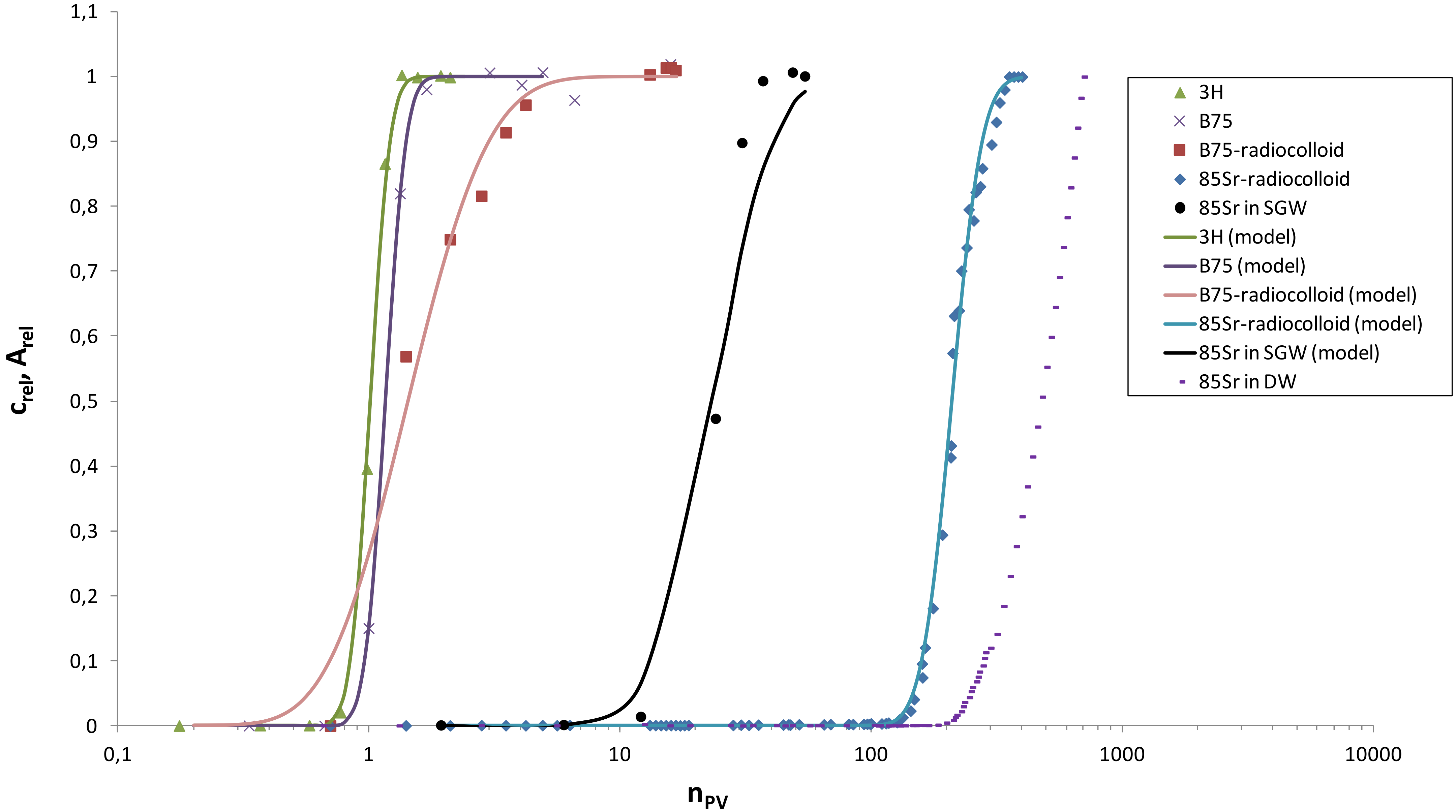


- sorption behavior
- 370 n_{PV}, 21 days
- the colloids passed column fast and left ^{85}Sr in column
- higher affinity of ^{85}Sr towards granite then colloids
- 10x slower ^{85}Sr transport in presence of colloids then ^{85}Sr transport in SGW without colloids

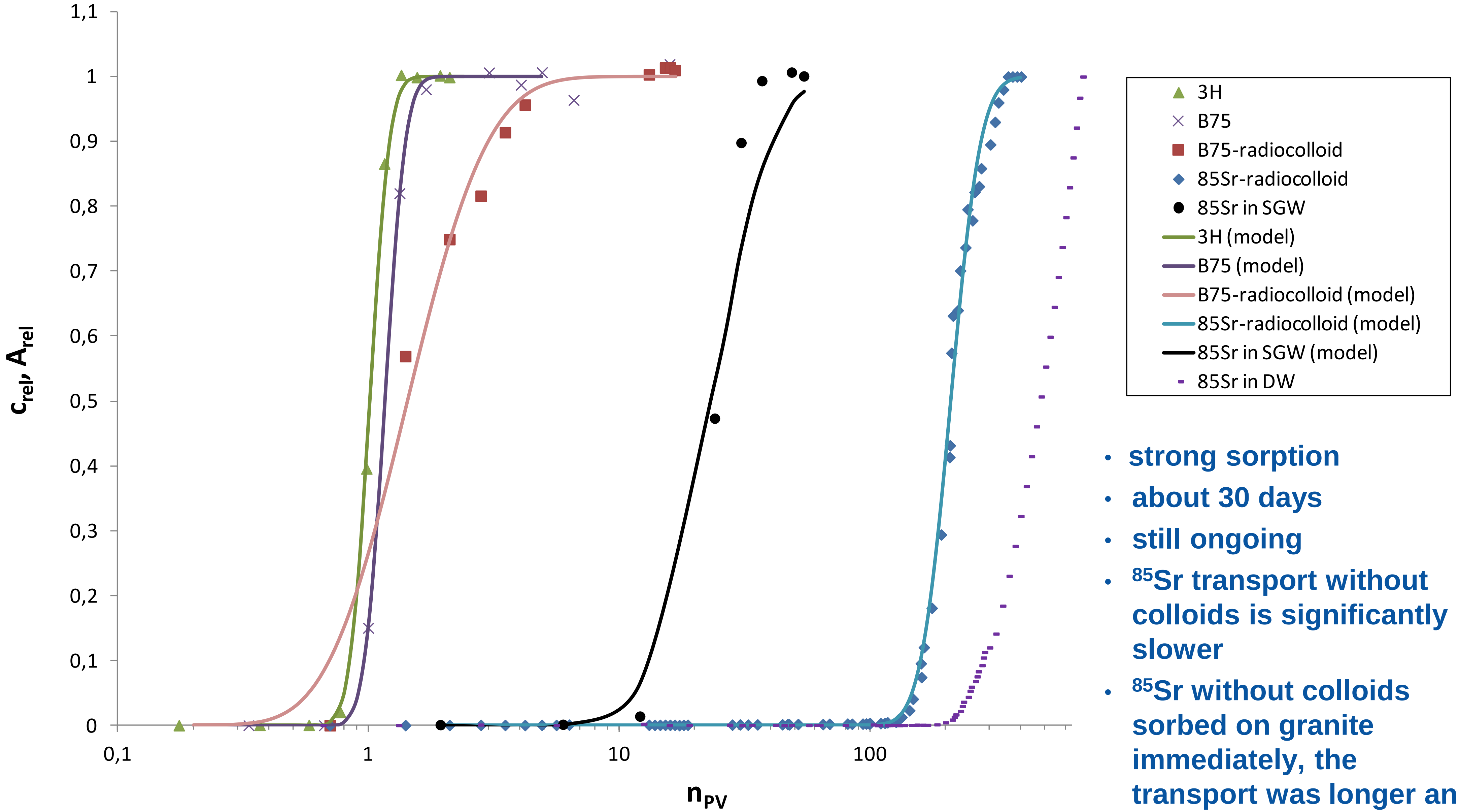
Dynamic experiments: ^{85}Sr (in DW)



Dynamic experiments: ^{85}Sr (in DW)

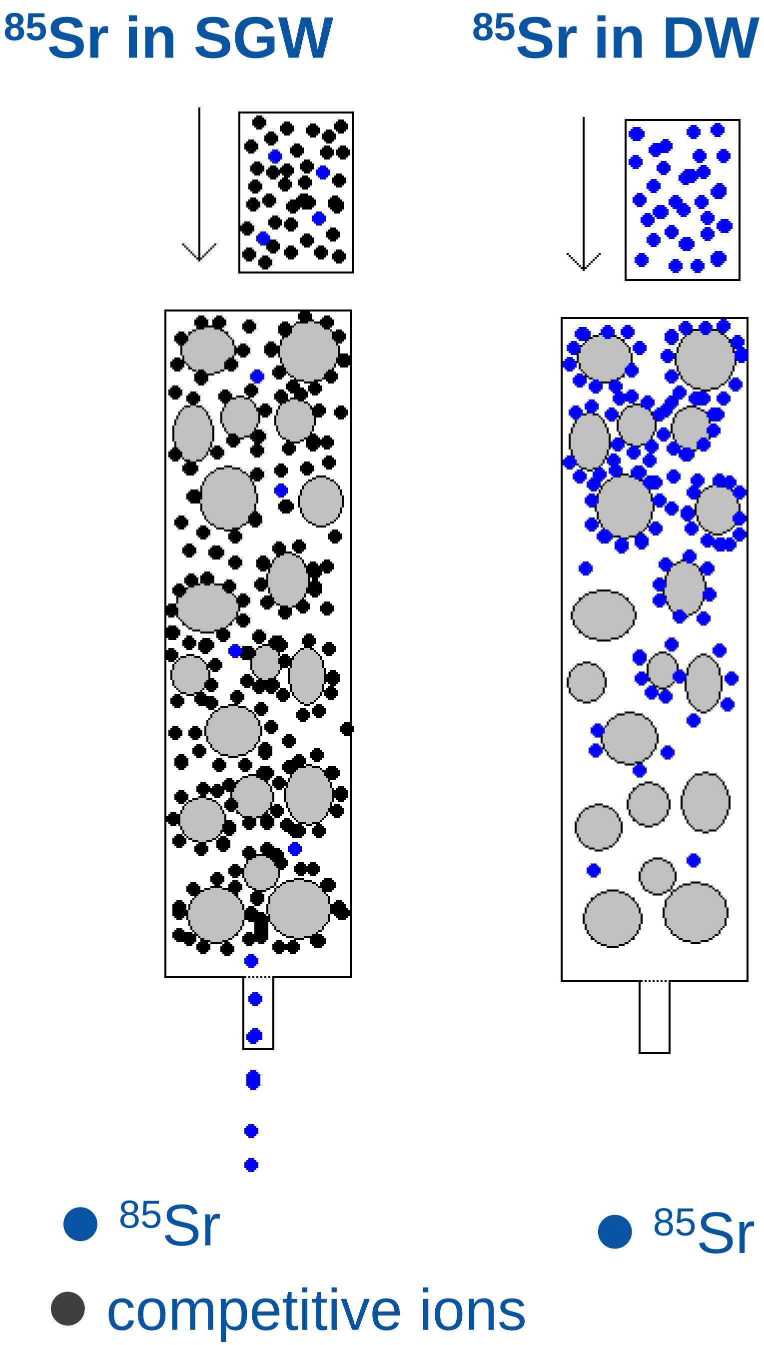


Dynamic experiments: ^{85}Sr (in DW)



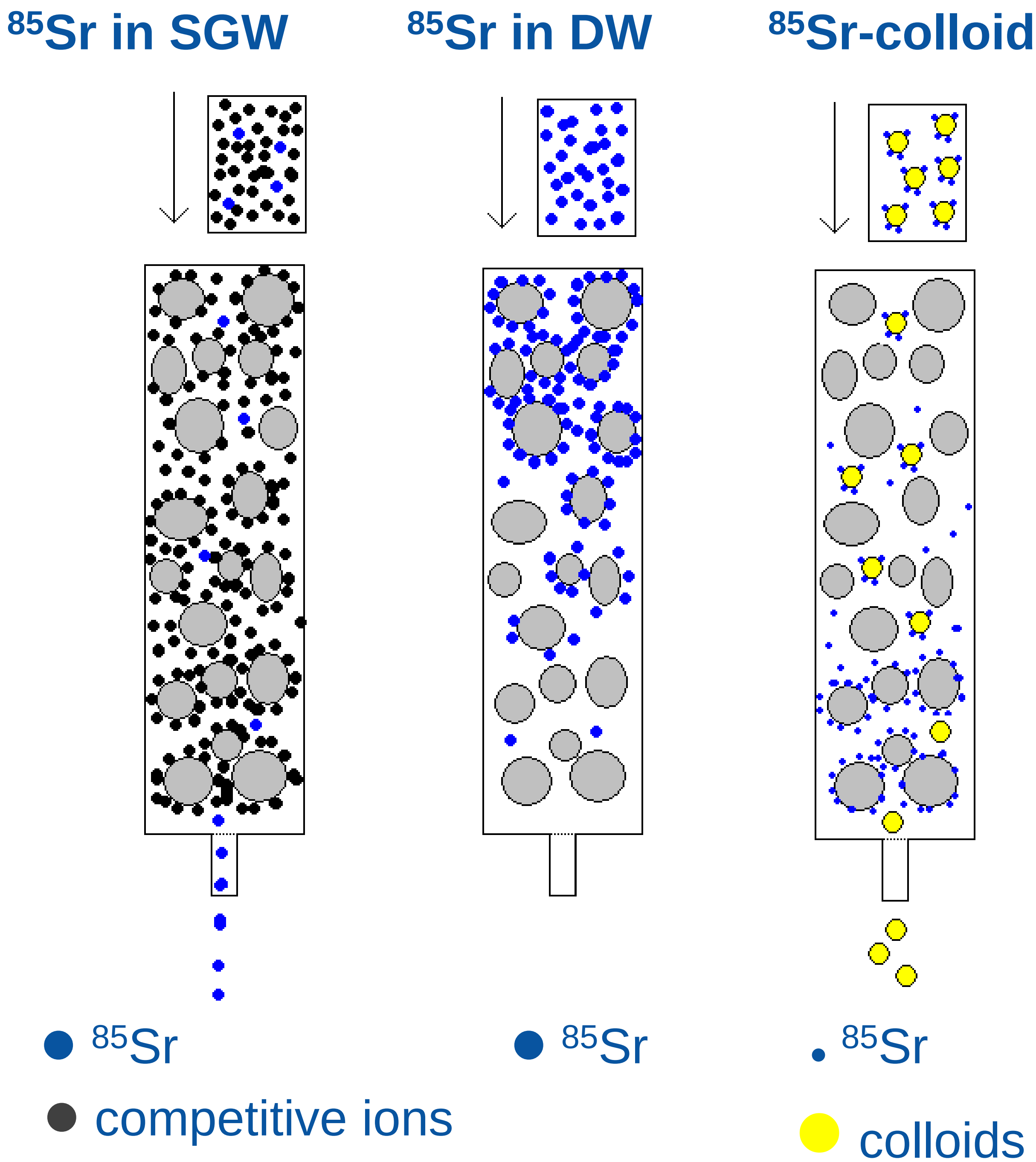
- strong sorption
- about 30 days
- still ongoing
- ^{85}Sr transport without colloids is significantly slower
- ^{85}Sr without colloids sorbed on granite immediately, the transport was longer and influenced by dispersion

Summary



- stronger ^{85}Sr sorption in DW
- significantly slower ^{85}Sr transport in DW then in SGW

Summary



- stronger ^{85}Sr sorption in DW
- significantly slower ^{85}Sr transport in DW then in SGW
- competitive effect of other ions in SGW
- ^{85}Sr transport without colloids is significantly slower
- ^{85}Sr on colloids passed in column further and after that is sorbed on granite
- ^{85}Sr without colloids sorbed on granite immediately, the transport was longer

Conclusion

Sorption experiments:

- ^{85}Sr sorption on granite is higher in presence of deionized water then in synthetic granitic water
- the mica minerals in granitic rock are not dominant sorbent of ^{85}Sr

Column experiments:

- Bentonite colloids transport:
  - bentonite colloids behaves as non-sorbing conservative tracer ^{3}H
  - transport of bentonite colloids through granite in presence of ^{85}Sr is slightly slower than transport of „pure“ colloids
- ^{85}Sr transport:
  - ^{85}Sr transport through granite in SGW is significantly faster than ^{85}Sr transport in DW
- Radiocolloids transport:
  - ^{85}Sr affinity is higher towards granite than towards bentonite colloids
  - ^{85}Sr transport through granite in presence of bentonite colloids in DW is faster then ^{85}Sr transport in DW

Acknowledgement

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no295487, the BELBaR project.