Synthesis and characterisation of scintillating microspheres made of polystyrene/polycarbonate for ^{222}Rn measurements

Pelay, E.; Tarancón, A.; Mitev, K.; Dutsov, Ch.; Georgiev, S.; Boshkova, T.; García, J.F.

Background

The absorption of ^{222}Rn in polymer samples can be described by the diffusion equation taking into account the radioactive decay:

$$\frac{\partial n(x,y,z,t)}{\partial t} = D\left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} + \frac{\partial^2 n}{\partial z^2}\right) - \lambda n$$

The solution for spherical specimen exposed to ambient concentration C_{env} for exposure time T_s and left to desorb in radon-free air is:

$$A_d(t) = 6\lambda K V C_{env} (L_D/R)^2 \sum_{j=1}^{\infty} \frac{1 - e^{-\lambda_j T_s}}{\lambda_j} e^{-\lambda_j t}$$

with: $$\lambda_j = \lambda\left(1 + \left(\frac{j\pi L_D}{R}\right)^2\right)$$

L_D and K are the physical parameters which describe the ^{222}Rn absorption in the polymer. By definition:

$$L_D = \sqrt{\frac{D}{\lambda}} \qquad K = \frac{C_{PC,0+}}{C_{env}}$$

Nuclear Instruments and Methods in Physics Research A 598 (2009) 620–627

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Sorption and desorption of radioactive noble gases in polycarbonates

D. Pressyanov *, K. Mitev, S. Georgiev, I. Dimitrova

Experimental Determination of L_D and K

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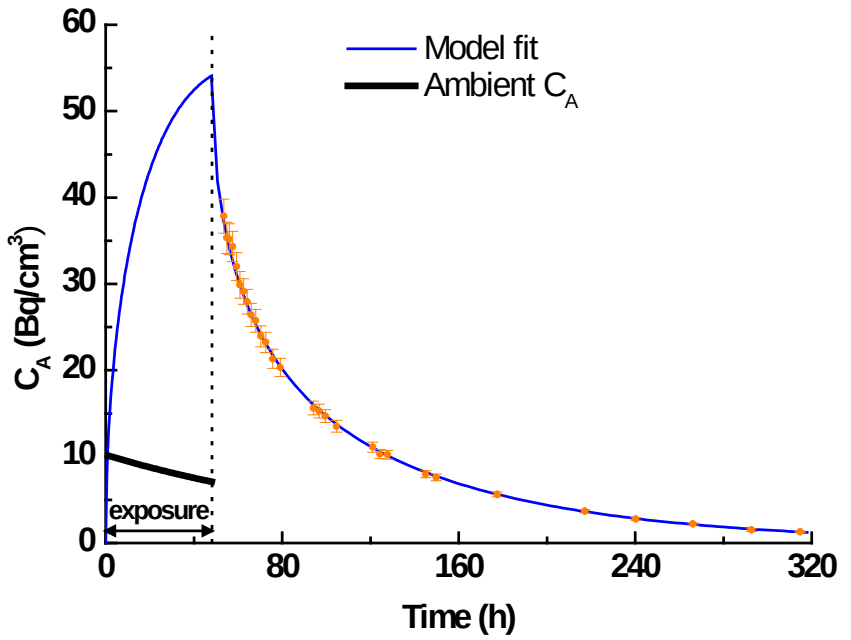
DETERMINATION OF THE DIFFUSION COEFFICIENT AND SOLUBILITY OF RADON IN PLASTICS

D. Pressyanov*, S. Georgiev, I. Dimitrova, K. Mitev and T. Boshkova

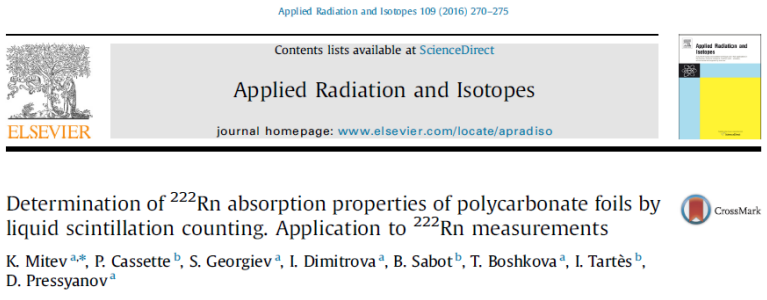


^{222}Rn, polymers and motivation

- **POLYCARBONATE**

  High ^{222}Rn absorption capabilities

  Track-etch properties


Applied Radiation and Isotopes 109 (2016) 270–275

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Determination of ^{222}Rn absorption properties of polycarbonate foils by liquid scintillation counting. Application to ^{222}Rn measurements

K. Mitev [a,*], P. Cassette [b], S. Georgiev [a], I. Dimitrova [a], B. Sabot [b], T. Boshkova [a], I. Tartès [b], D. Pressyanov [a]

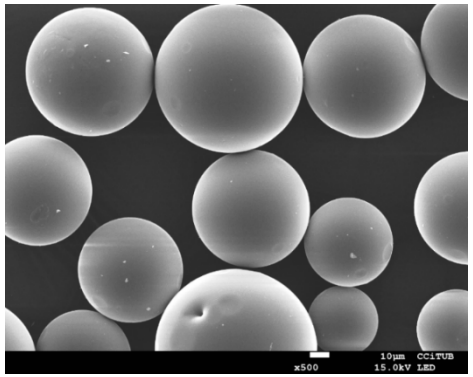
Material	Diffusion length, L_D (μm)	Partition coefficient from air, K
Makrofol® N	38.9 (13)	112 (12)
Makrofol® DE	52.1 (10)	26.2 (25)

[1]The values are for 20 ˚C.

- **PLASTIC SCINTILLATION MICROSPHERES MADE OF POLYSTYRENE**

  High scintillating capabilities

  Preliminary and successfully tested for ^{222}Rn absorption and measurement



IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 63, NO. 2, APRIL 2016 1209

Pilot Study of the Application of Plastic Scintillation Microspheres to Rn-222 Detection and Measurement

Krasimir Mitev, *Member, IEEE*, Ivelina Dimitrova, Alex Tarancón, Dobromir Pressyanov, Ludmil Tsankov, Tatiana Boshkova, Strahil Georgiev, Rositsa Sekalova, and Jose F. García

Material	Diffusion length, L_D (μm)	Partition coefficient from air, K
PSm from polystyrene	120.8 (21)	6.62(51)

[1]The values are for 21 ˚C.

Objectives

- Develop PSm made of Polystyrene, Polycarbonate and mixtures for the ^{222}Rn absorption and measurement.

✓ Synthesis of PSm of PS, PC and mixtures of PS and PC.

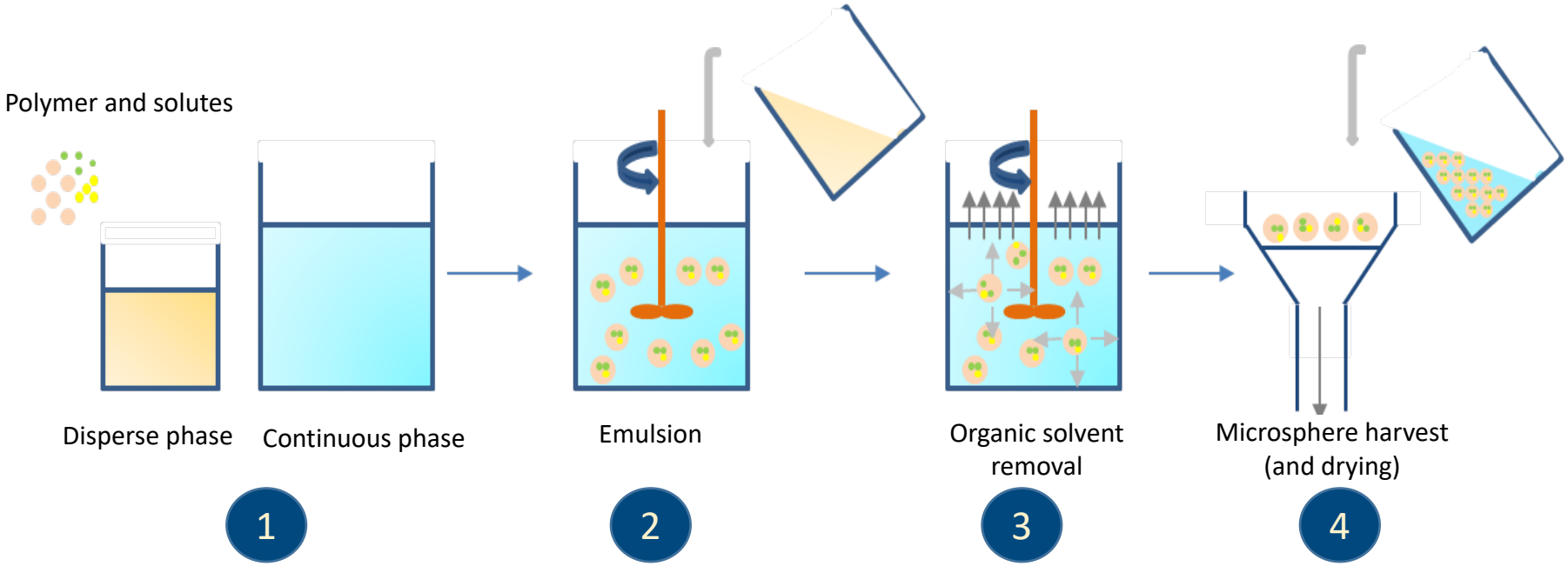
✓ Characterization (morphological and radiological) of the microspheres synthesised.

✓ Test the applicability of the PSm to measure ^{222}Rn.

Preparation of PSm

- **EVAPORATION/EXTRACTION METHOD**

1. Preparation of the Organic and Aqueous phase
2. Slow addition of phases using separation funnel
3. Stirring 20h at 25ºC
4. PSm filtered and cleaned with ethanol and water
5. Dried 12h at 40ºC



ORGANIC PHASE

- 25 g Polymer
- 0.2 % (w/w) PPO
- 0.05 % (w/w) POPOP
- 250 mL Dichloromethane

AQUEOUS PHASE

- 2 L Deionized water
- 1 % w/v PVA

Preparation of PSm

Raw materials

PS_SA
Polystyrene Across
260000 MW (average)

PC_MAKD
Polycarbonate
Makrofol D (Bayer)

PSm prepared

PSm	Polymer	Composition
1	PS_SA	25 g of polystyrene Across
2	PS_SA/PC_MAKD 75:25	18.75 g of Across polystyrene + 6.25 g of MAKD polycarbonate
3	PS_SA/PC_MAKD 50:50	12.5g of Across polystyrene + 12.5 g of MAKD polycarbonate
4	PS_SA/PC_MAKD 25:75	6,25 g of Across polystyrene +18.75 g of MAKD polycarbonate
5	PC_MAKD	25 g of MAKD polycarbonate

Preparation of PSm

PSm appearance

- 75g of PSm were prepared (3*25g).
- In all cases particles were obtained and efficiency was close to 100%.

Fine dust

PS_SA, PC_MAKD, PS/PC 50:50

Blocks

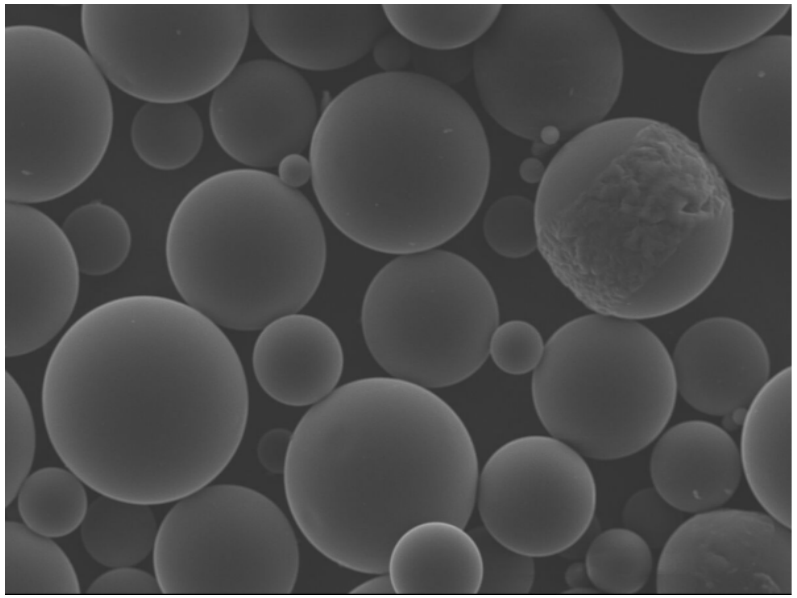
PS/PC 75:25

Agglomerates

PS/PC 25:75

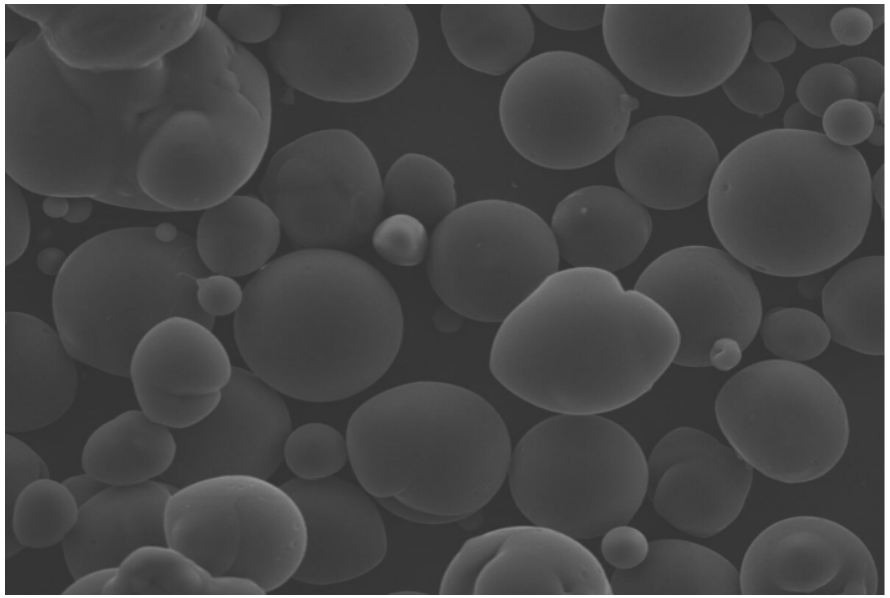
Preparation of PSm

PSm_SA (polystyrene)



- Median size 85µm
- Smooth
- Spherical without deformities

PSm_MAKD (polycarbonate)

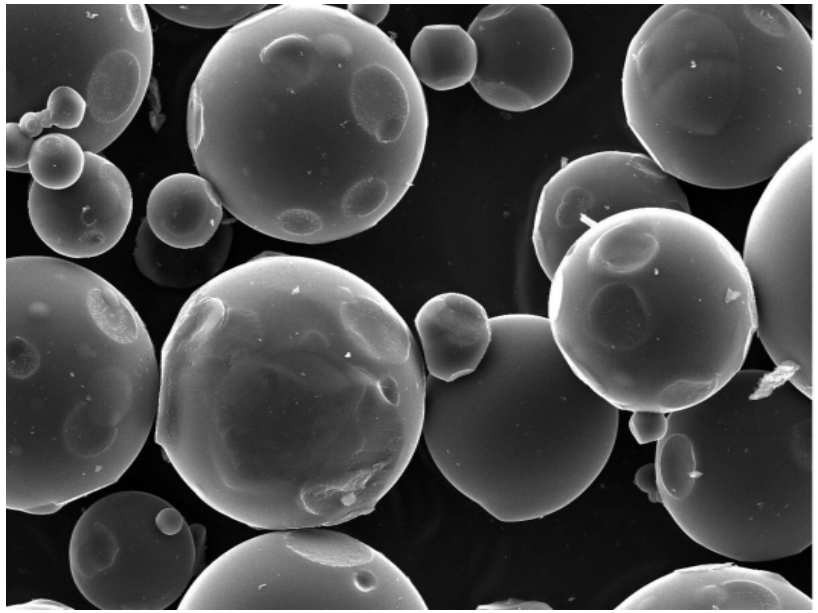


- Median size 65µm
- Smooth with Superficial holes
- Non-Spherical

✓ The PC PSm non-spherical form may be caused by the different formation of microdroplets and the process of solidification due to polarity of PC

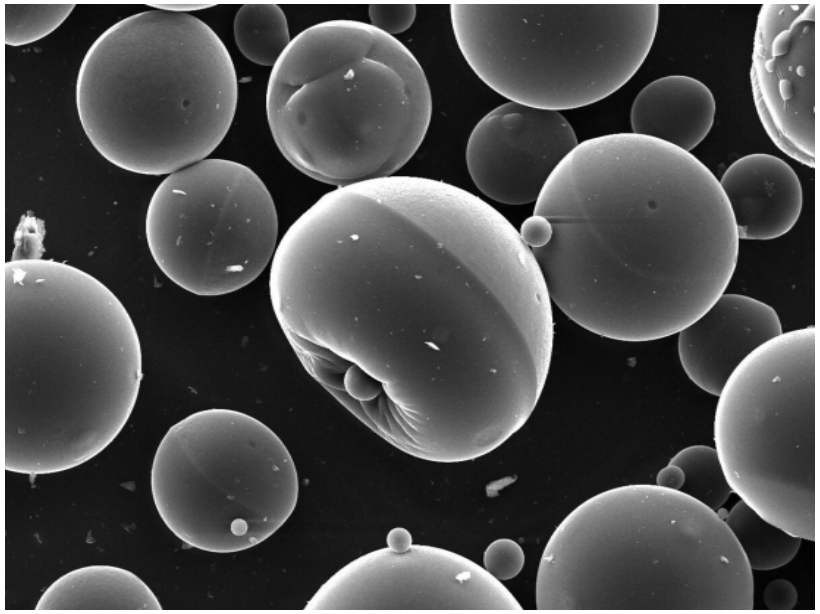
Preparation of PSm

PS/PC (75/25)



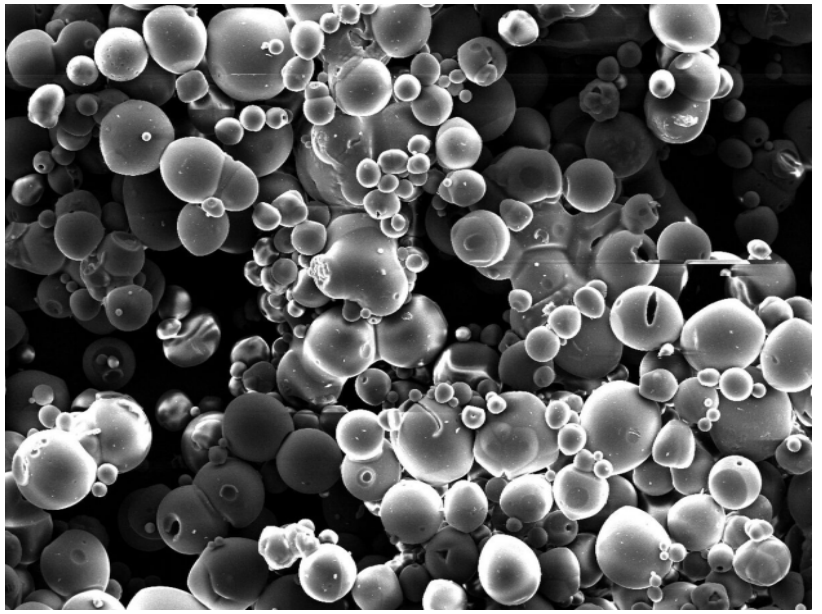
- Agglomerations
- Median size 65µm
- Plains formed in collisions

PS/PC (50/50)



- Median size 55 µm
- Non-spherical
- Line in the half

PS/PC (25/75)

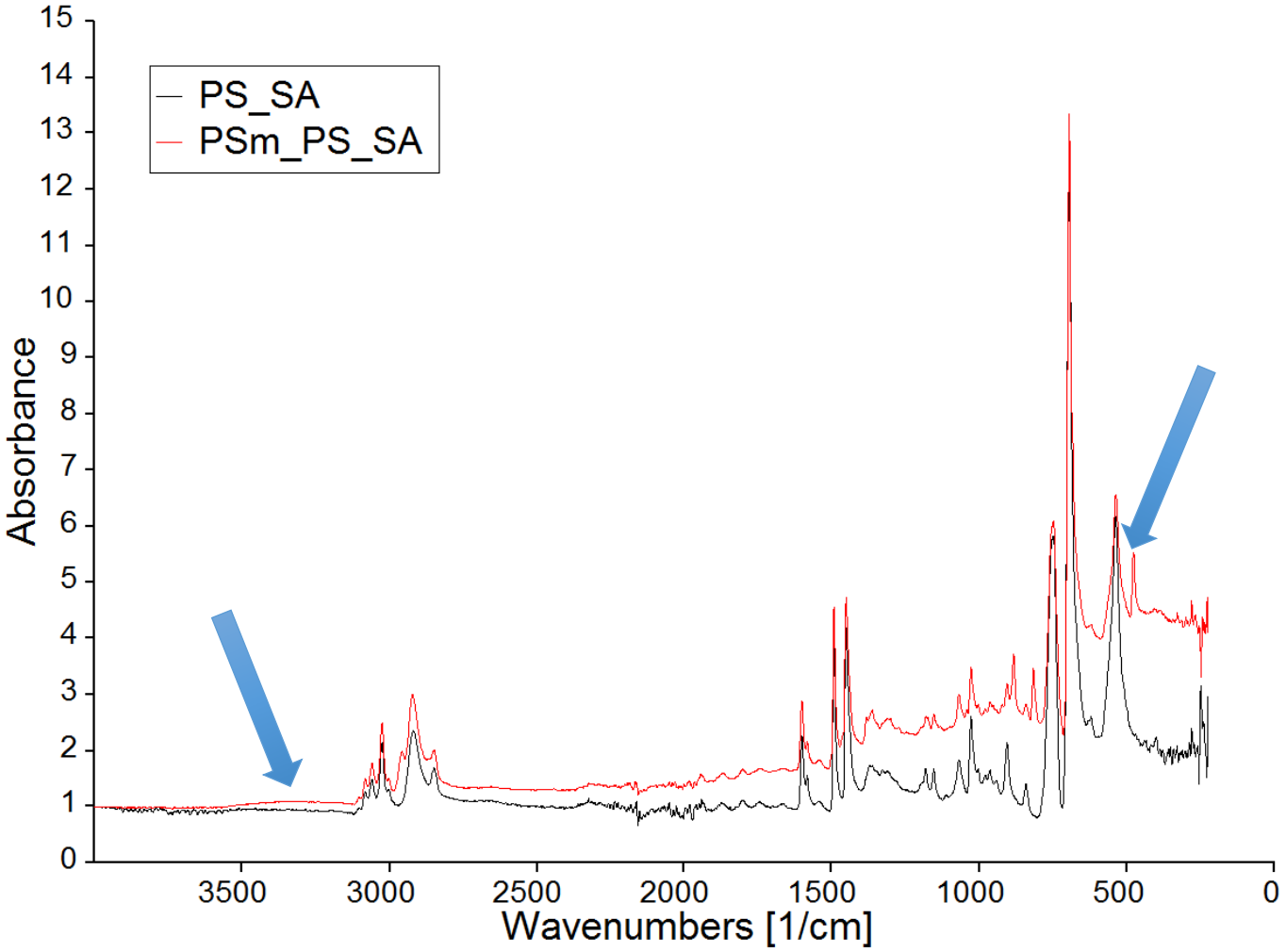


- Very big agglomerations
- Median size 100 µm
- Big holes

✓ PC and PS are not miscible and form two phases into the microdroplet before precipitate. This behaviour and its proportion defines the final shape

Preparation of PSm

IR analysis



- PS_SA (polystyrene)
- PSm_PS (microspheres of PS)

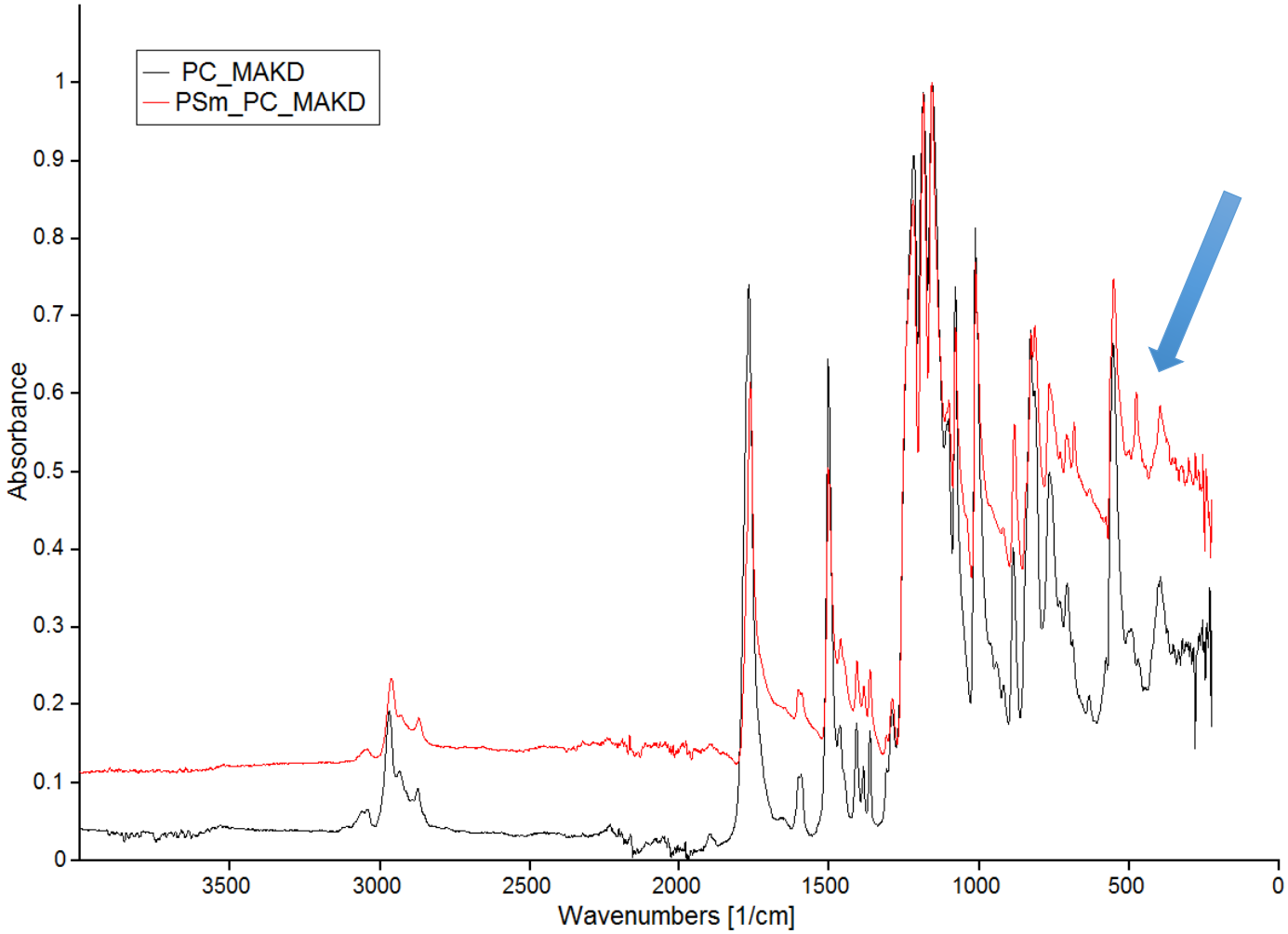
✓ Same peaks (PS is not modified)

✓ Encapsulation of solutes

✓ Some water and/or PVA?

Preparation of PSm

PSm morphology (IR analysis)



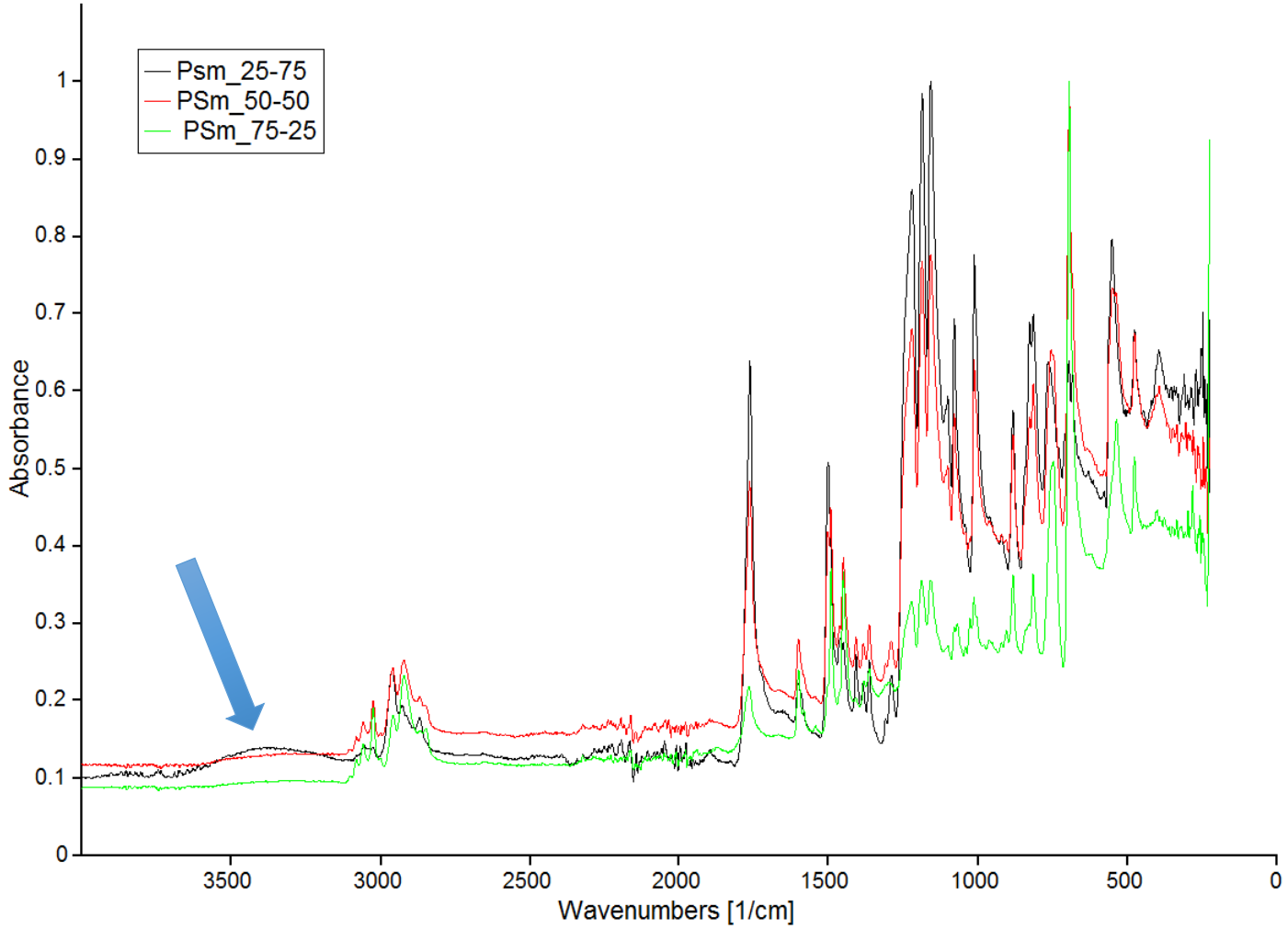
- PC_MAKD (polycarbonate)
- PSm_MAKD (microspheres of PC)

✓ Same peaks (PC is not modified)

✓ Encapsulation of solutes

Preparation of PSm

PSm morphology (IR analysis)



- PSm_PS_PC_75/25
- PSm_PS_PC_50/50
- PSm_PS_PC_25/75

✓ Same peaks (PC and PS are not modified)

✓ Encapsulation of solutes

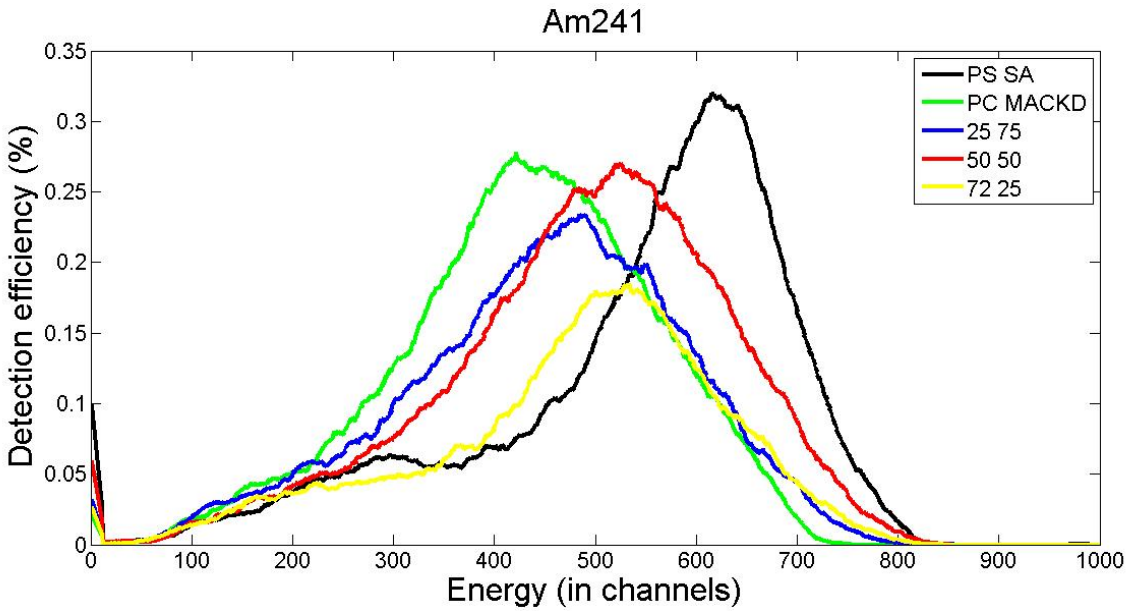
✓ Some PVA or water?

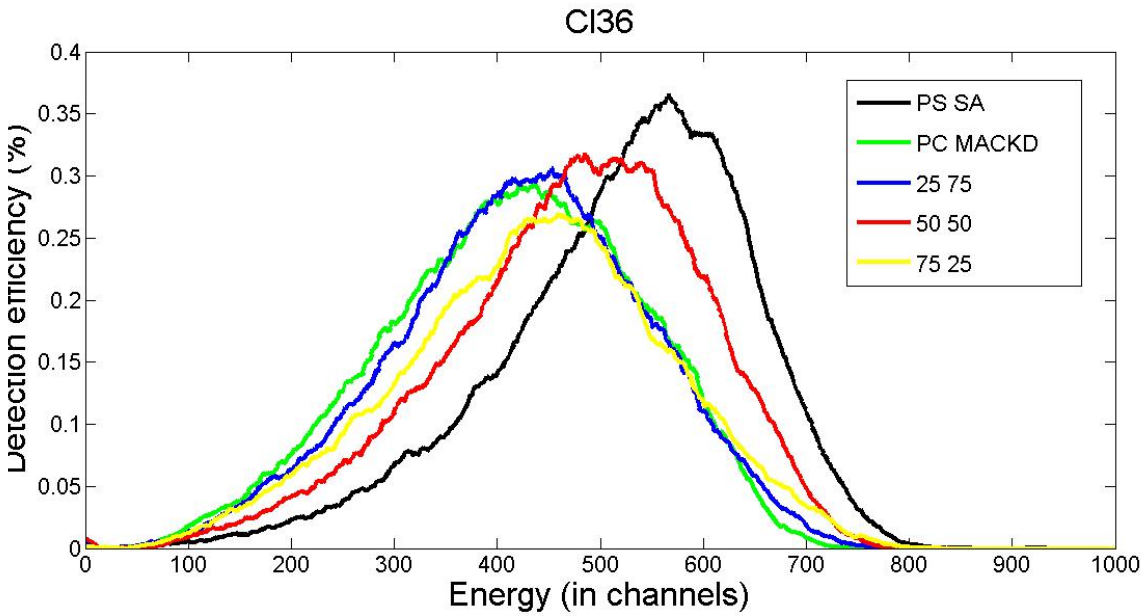
Radiometric Capabilities

Quantulus detector

Spectrum position and Quenching

	PS_PC (100/0)	PS_PC (75/25)	PS_PC (50/50)	PS_PC (25/75)	PS_PC (0/100)
SQP [E]	806(3)	799(4)	765(4)	745(5)	712(3)





✓ Quenching increases with the increase of the amount of PC (chemical quenching)

Radiometric Capabilities

Quantulus detector

Detection efficiency for high-energy beta and alpha emitters

	PS_PC (100/0)	PS_PC (75/25)	PS_PC (50/50)	PS_PC (25/75)	PS_PC (0/100)
Blank [cpm]	0.94(0.10)	0.75(0.08)	0.87(0.21)	0.87(0.27)	0.83(0.11)
SQP [E]	806(3)	799(4)	765(4)	745(5)	712(3)
Eff. ^{3}H [%]	**0.9(0.1)**	**0.30(0.03)**	**0.66(0.03)**	**0.47(0.03)**	**0.53(0.02)**
Eff. ^{36}Cl [%]	**97(1)**	**84(3)**	**96(1)**	**91(2)**	**93(1)**
Eff. ^{241}Am [%]	**75(1)**	**53(4)**	**85(2)**	**74(1)**	**77(1)**

- ✓ Detection efficiency for tritium is low
- ✓ Values are high in all cases except for 75/25 (agglomerations of PSm)
- ✓ Detection efficiency is mainly dependent of the PSm diameter.

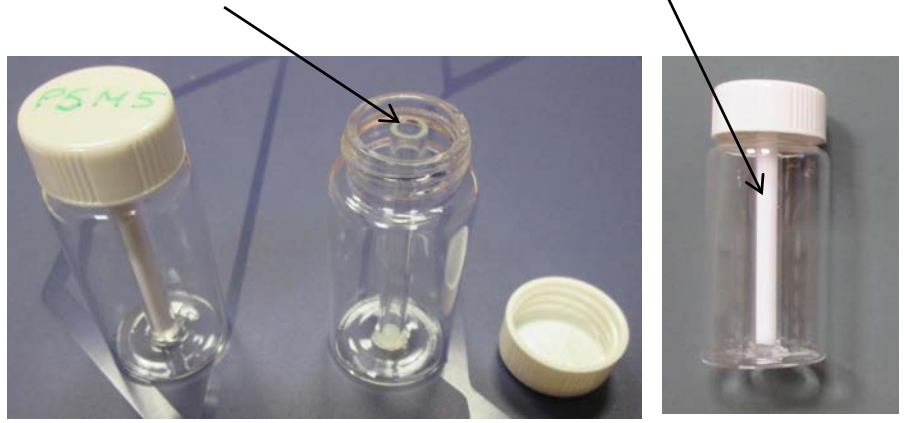
Radiometric Capabilities

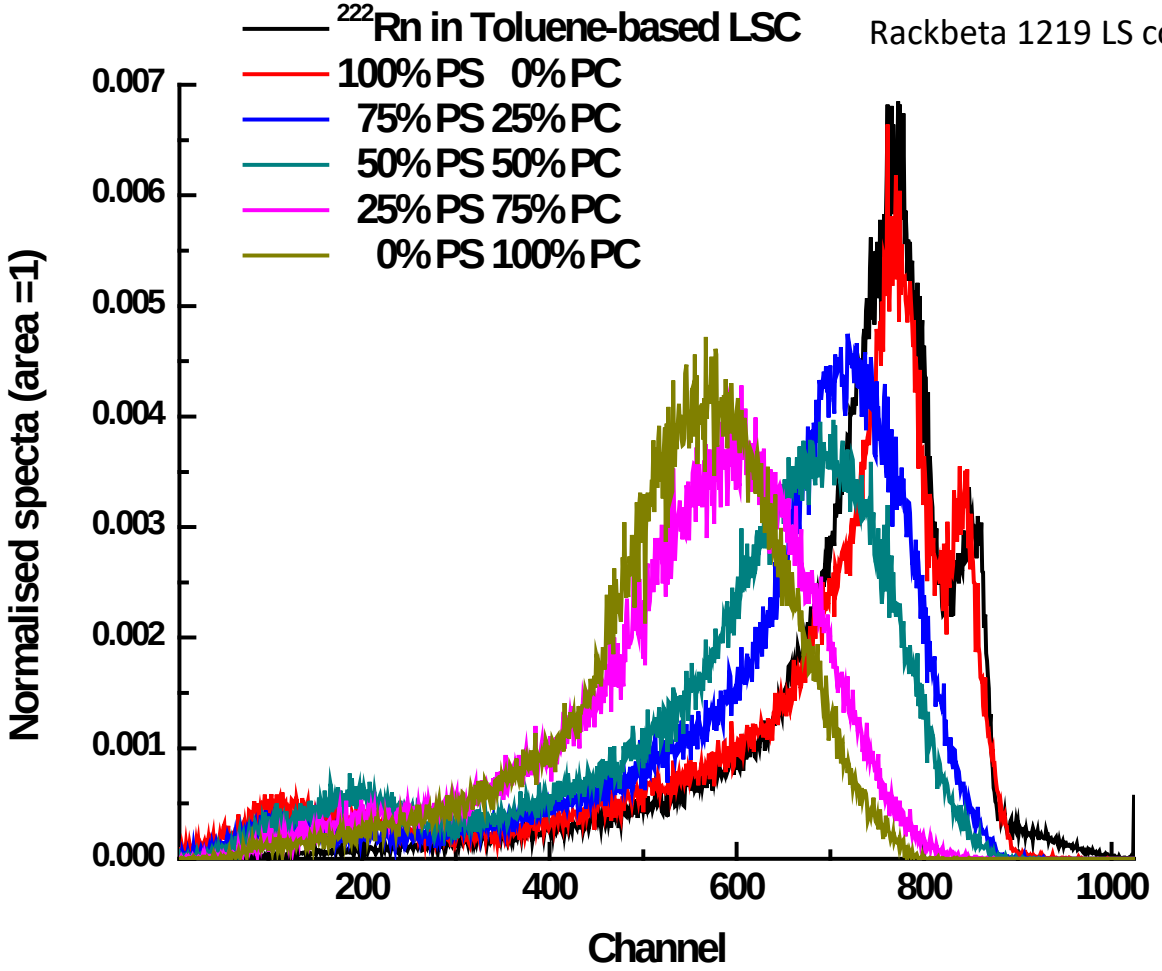
Spectra of ^{222}Rn absorbed in PSm (in equilibrium with its progeny)

Counting geometry optimized for better light collection as suggested by Philippe Cassette

Glass tube 5mm diameter

PSm





- ✓ Spectrum of PSm made of 100% polystyrene (PS) very similar to that of the LS cocktail
- ✓ Quenching increases with the increase of the polycarbonate content

^{222}Rn absorption properties

Partition coefficients and diffusion lengths

PSm	*K*
100% PS 0% PC	5.8 (15)
75% PS 25% PC	6.6 (17)
50% PS 50% PC	6.8 (17)
25% PS 75% PC	5.5 (14)
0% PS 100% PC	5.2 (13)

Compare to *K of the raw material*: 6.62(51)

26.2(25)

PSm	L_D, μm
100% PS 0% PC	97.0 (23)
75% PS 25% PC	69.4 (18)
50% PS 50% PC	49 (14)
25% PS 75% PC	1125 (45)
0% PS 100% PC	112.9 (46)

Compare to L_D: 120.8(31)

Residual PVA? 52.1(10)

Error introduced by non-spherical PSm?

- ✓ The production of PSm of polycarbobnate changed the radon absorption properties
- ✓ The change is more pronounced for the partition coefficient *K* and less pronounced for L_D

Conclusions

- ✓ PSm of PS, PC and mixtures of PS/PC can been prepared by the evaporation/extraction method
- ✓ Different polarity of PS and PC cause formation of PSm of different shape: spherical for PS and pseudo-spherical for PC.
- ✓ PC and PS are not miscible and in PSm of both are segregated.
- ✓ Efficiency of PSm of PC is slightly lower due to quenching effect caused by PC.
- ✓ The ^{222}Rn absorption properties of PSm made from 100% polycarbonate differ from the ^{222}Rn absorption properties of the polycarbonate material.

Acknowledgment

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Thank you for your attention.