

# Remark on the potential influence of the way of polymer production on their radon absorption properties

**Krasimir Mitev** 

Workshop: Transport of Radon and Thoron in Polymers

21-22 MARCH 2019, Faculty of Physics, Sofia University "St. Kliment Ohridski"

#### Note:

• This presentation contains more questions than answers.

## Gas diffusion in polymers

JOURNAL OF APPLIED PHYSICS

VOLUME 34, NUMBER 1

JANUARY 1963

#### Diffusion of Gases in Polyethylene Terephthalate

Alan S. Michaels, Wolf R. Vieth, and James A. Barrie\*

Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts

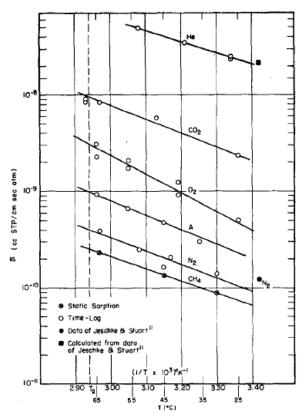


Fig. 1. Permeability constants in amorphous polyethylene terephthalate.

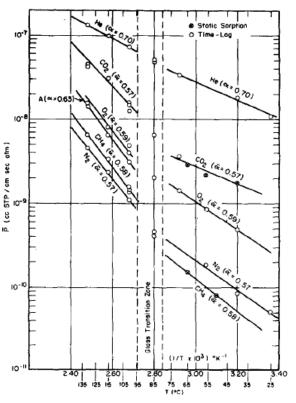


Fig. 2. Permeability constants in crystalline polyethylene terephthalate.

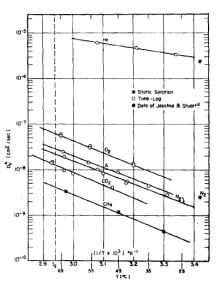


Fig. 3. Diffusion constants in amorphous polyethylene terephthalate.

#### E. Correlation of Diffusion Constants in the Crystalline Polymer with Gas Molecular Size

Since apparent activation energies correlate with  $d^2$  in both the glassy and rubbery states and Eq. (4) is obeyed, it is logical to assume that  $\ln D$  will correlate with  $d^2$  in either state of the polymer. These correlations are successful, as shown in Fig. 6, within mean precision limits of  $\pm 40\%$  at the 95% confidence level. Equations of the type  $\ln D = A - bd^2$  are obtained. Equation (9) applies to the glassy polymer at  $40^{\circ}\text{C}$  while Eq. (10) applies to the rubbery crystalline polymer at  $130^{\circ}\text{C}$ .

$$\ln D = -12.2 - 0.51d^2 \qquad (d \text{ in Å}) \tag{9}$$

$$\ln D = -9.9 - 0.38d^2. \tag{10}$$

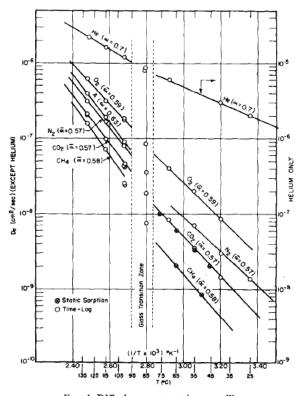


Fig. 4. Diffusion constants in crystalline polyethylene terephthalate.

# When we determined $L_D$ and K of Xe-131 and Kr-85 in Makrofol N and DE we discussed that with Philippe Cassette

Applied Radiation and Isotopes 134 (2018) 269-27



#### Contents lists available at ScienceDirect

#### Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso



Diffusion lengths and partition coefficients of  $^{131\rm m}$ Xe and  $^{85}$ Kr in Makrofol N and Makrofol DE polycarbonates

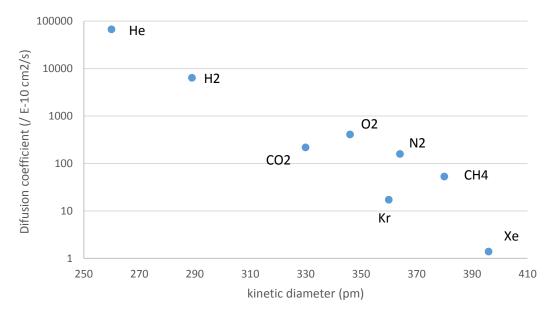


K. Miteva, P. Cassette, I. Tartès, S. Georgieva, I. Dimitrova, D. Pressyanova

<sup>a</sup> Faculty of Physics, Sofia University "St. Kliment Ohridski", 1164 Sofia, Bulgaria
<sup>b</sup> CEA, LIST, Laboratoire National Henri Becquerel, Gif-sur-Yvette, LNE-LNHB, F-91191, France

but also for the partition coefficient K. Thus, in terms of the free volume theory, our hypothesis is that both  $L_D$  and K depend on the free volume in the polycarbonate and the free volume is different for RNGs with different atomic radii. More in detail, our hypothesis is that the trapping of RNG atoms in volume traps available in the polycarbonate is of paramount importance for their absorption and transport in the material. This hypothesis implies enhanced Langmuir-type adsorption and suggests how it is possible to have K>1. In addition, this hypothesis explains why the diffusion coefficients of RNGs determined in this work (Table 8) are much smaller than the diffusion coefficients of other gases in polycarbonate. Notice that if we compare, for example, the Makrofol\* DE diffusion coefficients of  $^{85}$ Kr (D = 17.2 \*10<sup>-10</sup> cm<sup>2</sup>/s),  $^{131m}$ Xe (D = 1.412  $*10^{-10} \text{ cm}^2/\text{s}$ ) and  $^{222}\text{Rn}$  (D = 0.569  $*10^{-10} \text{ cm}^2/\text{s}$ ) determined here with the polycarbonate diffusion coefficients published by Thran2 et al. (1999) for He ( $D = 67300 \times 10^{-10} \text{ cm}^2/\text{s}$ ), H<sub>2</sub> ( $D = 6400 \times 10^{-10} \text{ cm}^2/\text{s}$ ),  $O_2(D = 411 *10^{-10} \text{ cm}^2/\text{s}), N_2(D = 159 *10^{-10} \text{ cm}^2/\text{s}), CO_2(D = 219)$  $*10^{-10} \text{ cm}^2/\text{s}$ ) and CH<sub>4</sub> (D = 53  $*10^{-10} \text{ cm}^2/\text{s}$ ) we observe between one or two orders of magnitude smaller diffusion coefficients for 131mXe and 222Rn. Evidently, there is something that impedes the RNG diffusion in the polycarbonate matrix and we suspect trapping of RNG atoms in volume traps.

#### Diffusion coefficient vs. Kinetic diameter



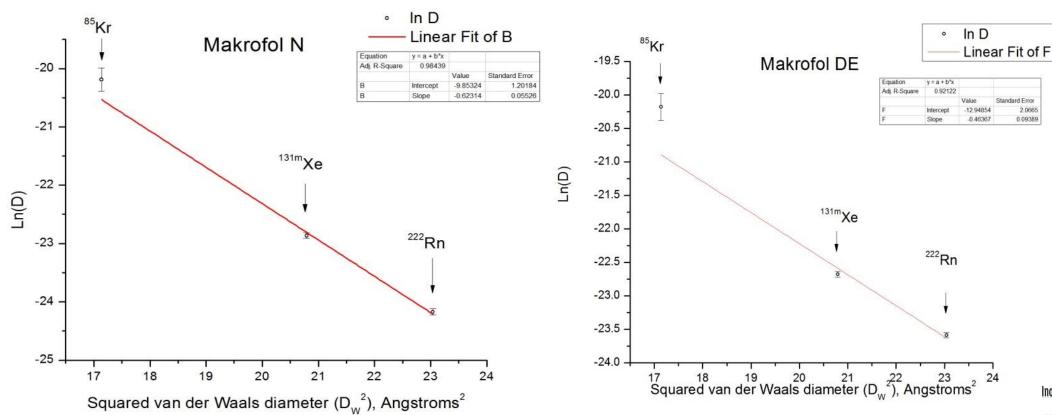
From P. Cassette, private communication

#### Dear Krasi,

Yes, the behavior of molecular gas is a little bit strange. I plotted in the attached file, the diffusion coefficient vs. the kinetic diameter of the gas (I do not know if this is the pertinent quantity and perhaps some kind of molecular diameter would be more appropriate). It seems that there is a nice exponential correlation (D is in log scale) for the molecular gas (except CO2) and also for noble gas but with another exponential constant. The two curves cross for H2. The behavior of CO2 is atypical.

#### D for noble gases in polycarbonates

	R	LDi	n Makrofol N	LD in Makr	ofol DE	l	Makrofol N			Makrofol D	E		Makrofol	N		Makrofol	DE
Nuclide	Angstrioms	LD	Sigma LD	LD	Sigma LD	LD, cm	D cm2/s	Sigma D	LD, cm	D cm2/S	Sigma D	<b>R2, A</b> <sup>2</sup>	ln D	Sigma Ln(D)	$R^2, A^2$	ln D	Sigma Ln(D)
<sup>85</sup> Kr	2.07	9140	910	9180	920	0.914	1.707E-09	3.398E-10	0.918	1.72159E-09	3.45068E-10	17.1396	-20.1888	1.991E-01	17.1396	-20.18	0.20043573
<sup>131m</sup> Xe	2.28	132.4	3.2	145.1	3.2	0.01324	1.176E-10	5.683E-12	0.01451	1.41203E-10	6.2281E-12	20.7936	-22.864	4.834E-02	20.7936	-22.6808	0.04410751
<sup>222</sup> Rn	2.4	38.9	1	52.1	1	0.00389	3.175E-11	1.633E-12	0.00521	5.69587E-11	2.18651E-12	23.04	-24.173	5.141E-02	23.04	-23.5887	0.03838772



# There are indications that gas transport in glassy polymers depend on the free volume in the polymers

THE JOURNAL OF CHEMICAL PHYSICS

VOLUME 31, NUMBER 5

NOVEMBER, 1959

#### Molecular Transport in Liquids and Glasses

MORREL H. COHEN, University of Chicago, Chicago, Illinois

David Turnbull, General Electric Research Laboratory, Schenectady, New York

(Received May 15, 1959)

We have derived, by using simple considerations, a relation between the diffusion constant D in a liquid of hard spheres and the "free volume"  $v_f$ . This derivation is based on the concept that statistical redistribution of the free volume occasionally opens up voids large enough for diffusive displacement. The relation is  $D = A \exp[-\gamma v^*/v_f]$ , where  $v^*$  is the minimum required volume of the void and A and  $\gamma$  are constants. This equation is of the same form as Doolittle's [J. Appl. Phys. 22, 1471 (1951)] empirical relation between the fluidity  $\phi$  of simple hydrocarbons and their free volume. It has been shown [Williams, Landel, and Ferry, J. Am. Chem. Soc. 77, 3701 (1955)] that the Doolittle equation also can be adapted to describe the abrupt decrease in molecular kinetic constants with decreasing temperature that accompanies the glass transition in certain liquids. Our result predicts that even the simplest liquids would go through this glass transition if sufficiently undercooled and crystallization did not occur. The problem of transport in actual simple and network liquids also is discussed.

It is shown that data on self-diffusion in some simple van der Waals liquids and liquid metals are described satisfactorily by our relation with v\* near the molecular volume for the van der Waals liquids and near the volume of the ion, corresponding to the highest valence state, for the metals.





#### Polymer Free Volume and Its Connection to the Glass Transition

Ronald P. White and Jane E. G. Lipson\*

Department of Chemistry, Dartmouth College, Hanover, New Hampshire 03755, United States

# Correlation Between Fractional Free Volume and Diffusivity of Gas Molecules in Glassy Polymers

A. THRAN, G. KROLL, F. FAUPEL

Technische Fakultät der Universität Kiel, Lehrstuhl für Materialverbunde, Kaiserstr. 2, D-24143 Kiel, Germany



Available online at www.sciencedirect.com

Journal of Membrane Science 226 (2003) 51-61



www.elsevier.com/locate/memsci

#### Effect of free volume and sorption on membrane gas transport

#### Chien-Chieh Hu, Chin-Shun Chang, Ruoh-Chyu Ruaan\*, Juin-Yih Lai

Department of Chemical Engineering, Research and Development Center for Membrane Technology, Chung Yuan University, Chung-Li 320, Taiwan

Received 8 October 2002; received in revised form 25 July 2003; accepted 30 July 2003

Advances in Colloid and Interface Science 164 (2011) 89-99



Contents lists available at ScienceDirect

#### Advances in Colloid and Interface Science

journal homepage: www.elsevier.com/locate/cis



Polymeric membrane materials: New aspects of empirical approaches to prediction of gas permeability parameters in relation to permanent gases, linear lower hydrocarbons and some toxic gases

O.V. Malykh b, A.Yu. Golub a, V.V. Teplyakov a,b,\*

a A.V. Topchiev Institute of Petrochemical Synthesis, Russian Academy of Sciences, Moscow, Russia

b Lomonosov Moscow State University, Faculty of Chemistry, Department of Chemical Technology and New Materials, Moscow, Russia



Materials Science and Engineering A317 (2001) 70-76



Molecularly-based numerical evaluation of free volume in amorphous polymers

Santosh Putta, Sia Nemat-Nasser \*

Department of Applied Mechanics and Engineering Science, Center of Excellence for Advanced Materials, University of California, San Diego,
La Jolia, CA 92093-0416, USA

Macromolecules 2003, 36, 8673-8684

Effects of Cooling Rate and Physical Aging on the Gas Transport Properties in Polycarbonate

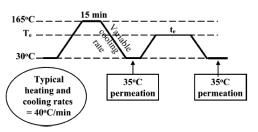
Christelle M. Laot,<sup>‡</sup> Eva Marand,<sup>\*</sup> B. Schmittmann,<sup>†</sup> and R. K. P. Zia<sup>†</sup>

Department of Chemical Engineering, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061-0211

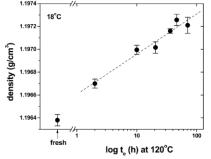
Received November 27, 2002

ABSTRACT: The gas transport properties of bisphenol A polycarbonate films were examined as a function of the cooling rate during film processing and subsequent physical aging. Interpretation of the permeation properties using results from dynamic mechanical analysis and density measurements indicated that the diffusion coefficient of small gas molecules in glassy polycarbonate is influenced more by the local chain dynamics rather than by the overall free volume content. A one-dimensional lattice model was developed to help probe the effects of the distribution of energy barriers associated with polymer motion on the transport properties. Fast cooling rates generated highly restricted conformations, which hindered local motions leading to increases in the measured activation energy of diffusion. These results correlated well with a lattice model having a broad distribution of energy barriers. Annealing reduced the measured diffusion coefficients, which was shown to be consistent with narrowing of the energy barrier distribution, as well as an increase in the mean barrier energy. Interestingly, the decrease in the diffusion coefficient with aging was found to occur much more slowly in fast-cooled samples, despite the higher initial free volume content. By contrast, properties such as density or isothermal dynamic mechanical properties were more consistent with the free volume model.

#### Production matters! Laot et. al.:



**Figure 2.** Schematic of the temperature program for the cooling rate studies. The polycarbonate films were heated from room temperature (30 °C) to 165 °C at a heating rate of 40 °C/min, kept at 165 °C for 15 min to remove thermal history, and cooled with the internal fan oven at several controlled rates to room temperature. The cooling rates were taken as 0.5 °C/min, 10 °C/min, 40 °C/min, or in excess of 40 °C/min (external fan).



**Figure 7.** Density as a function of the aging time for a sample cooled at 40 °C/min and aged at 120 °C. The densities were measured at 18 °C using a linear density gradient column.

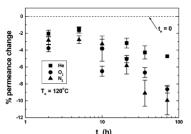
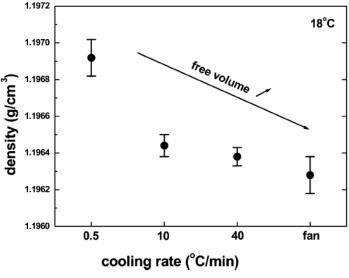
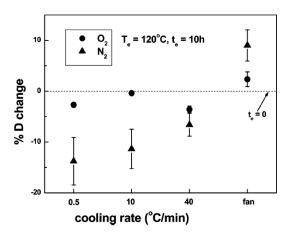


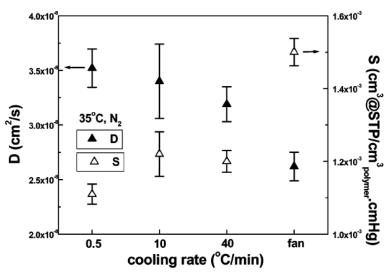
Figure 10. Percentage of the permeance change as a function of annealing time at 120 °C for the gases studied in this research. The permeation slopes were obtained at 35 °C and averaged. As apparent from the figure, the permeation slope was reduced with annealing time.



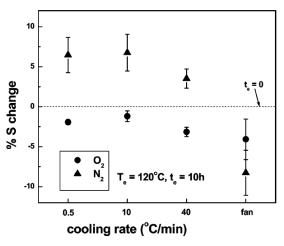
**Figure 3.** Density as a function of the cooling rate. The densities were measured at 18 °C using a linear density gradient column.



**Figure 16.** Percent in diffusion coefficient change as a function of the cooling rate for the various gases of interest. The samples had been annealed at 120  $^{\circ}$ C for 10 h. The permeation experiments were carried out at 35  $^{\circ}$ C.



**Figure 4.** Diffusion coefficient D and solubility coefficient S for  $N_2$  as a function of cooling rate. The permeation experiments were performed at 35 °C. The applied pressure was fixed at about 3.9 atm for  $N_2$ .



**Figure 17.** Percent in solubility coefficient change as a function of the cooling rate for the various gases of interest. The samples had been annealed at  $120~^{\circ}\text{C}$  for 10~h. The permeation experiments were carried out at  $35~^{\circ}\text{C}$ .

Fluid Phase Equilibria 444 (2017) 47–55



Contents lists available at ScienceDirect

#### Fluid Phase Equilibria

journal homepage: www.elsevier.com/locate/fluid



# Predictive model for gas and vapor sorption and swelling in glassy polymers: II. Effect of sample previous history



Matteo Minelli\*, Ferruccio Doghieri

Department of Civil, Chemical, Environmental and Materials Engineering — DICAM, Alma Mater Studiorum, University of Bologna, Via Terracini 28, I-40131, Bologna, Italy

#### ARTICLEINFO

Article history:
Received 15 February 2017
Received in revised form
21 April 2017
Accepted 22 April 2017
Available online 25 April 2017

Keywords: Fluid solubility Thermodynamics Glassy polymers NELF model Polymer swelling

#### ABSTRACT

The solubility of various gases in several glassy polymers has been analyzed by the nonequilibrium thermodynamics for glassy polymers (NET-GP) framework, coupled with a lattice fluid equation of state model. Moreover, a simple rheological tool recently introduced for the a priori evaluation of the penetrant induced swelling is employed to predict the volumetric behavior of the solute/polymer mixture below T<sub>g</sub>. Such model accounts for an additional nonequilibrium parameter for the description of polymer phase that represents its pseudoequilibrium compressibility, and it can be retrieved from pure component pressure—volume—temperature data below T<sub>g</sub>. More in detail, the effect of polymer pretreatment and its prior history (e.g. annealing or conditioning by preswelling) on the resulting solubility behavior has been thoroughly analyzed and discussed, and the experimental data obtained for several penetrant/polymer couples have been well described by the present model.

© 2017 Elsevier B.V. All rights reserved.

#### **Production matters!**

#### 4. Conclusions

The solubility of various penetrants in glassy polymers has been analyzed and described by means of the nonequilibrium thermodynamics for glassy polymers (NET-GP) approach, which has been used in combination with the lattice fluid EoS by Sanchez and Lacombe. The resulting nonequilibrium lattice fluid model (NELF), coupled with the novel rheological approach for the prediction of solute induced swelling, provides the a priori evaluation of the solubility of any penetrant in polymer systems below their  $T_g$ , even in case a significant swelling is induced by the solute.

In this work, a particular care has been devoted to the analysis of the effect of sample pretreatment on the resulting solubility isotherms. Indeed, various previous histories of the polymer glass (e.g. annealing, conditioning, pre-swelling etc.) often lead to significant differences in the resulting penetrant solubility. The NELF model accounts for the diverse initial nonequilibrium states of the polymer glass assuming different values of the initial density, and the

#### **Comment**

My primary concern here is about the fact that we are using polycarbonates like Makrofol DE and Makrofol N\*, but we have little or no information about the production history of these polymers.

\* The usage of Makrofol N foil for radon studies was suggested and kindly provided by Luigi Tommasino. The usefulness of this support is very difficult to exaggerate. Thank you!

J Radioanal Nucl Chem (2017) 314:637–649 DOI 10.1007/s10967-017-5488-3



# Synthesis and characterisation of scintillating microspheres made of polystyrene/polycarbonate for <sup>222</sup>Rn measurements

Eduard Pelay<sup>1</sup> · Alex Tarancón<sup>1,3</sup> · Krasimir Mitev<sup>2</sup> · Chavdar Dutsov<sup>2</sup> · Strahil Georgiev<sup>2</sup> · Ludmil Tsankov<sup>2</sup> · José Francisco García<sup>1</sup>







# Synthesis and characterisation of scintillating microspheres made of polystyrene/polycarbonate for <sup>222</sup>Rn measurements

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

## Background

The absorption of <sup>222</sup>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:

$$A_d(t) = 6\lambda K V C_{env} \left( L_D/R \right)^2 \sum_{j=1}^{\infty} \frac{1 - e^{-\lambda_j T_s}}{\lambda_j} e^{-\lambda_j t}$$
 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 <sup>222</sup>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



Contents lists available at ScienceDirect

#### Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima



Sorption and desorption of radioactive noble gases in polycarbonates

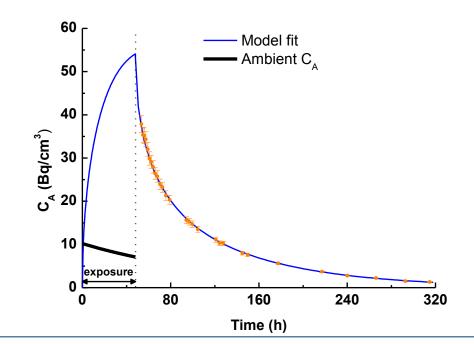
D. Pressyanov \*, K. Mitev, S. Georgiev, I. Dimitrova

#### Experimental Determination of $L_D$ and K

Radiation Protection Dosimetry (2011), Vol. 145, No. 2-3, pp. 123-126 Advance Access publication 5 April 2011 doi:10.1093/rpd/ncr069

#### DETERMINATION OF THE DIFFUSION COEFFICIENT AND SOLUBILITY OF RADON IN PLASTICS

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



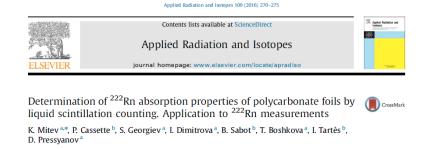
# <sup>222</sup>Rn, polymers and motivation

#### POLYCARBONATE

High <sup>222</sup>Rn absorption capabilities

Track-etch properties





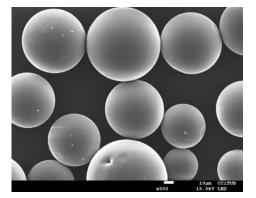
Material	Diffusion length, $L_D$ ( $\mu$ m)	Partition coefficient fron air, <i>K</i>	
Makrofol® N	38.9 (13)	112 (12)	
Makrofol® DE	52.1 (10)	26.2 (25)	

<sup>1</sup>The values are for 20 °C.

#### PLASTIC SCINTILLATION MICROSPHERES MADE OF POLYSTYRENE

High scintillating capabilities

Preliminary and successfully tested for <sup>222</sup>Rn absorption and measurement



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

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

	Diffusion	Partition	
Material	length, $L_D$	coefficient	
	(µm)	from air, K	
PSm from	120.8 (21)	6.62(51)	
polystyrene	1Tho x	volues are for 21 °C	

<sup>1</sup>The values are for 21 °C.

## **Objectives**

• Develop PSm made of Polystyrene, Polycarbonate and mixtures for the <sup>222</sup>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 <sup>222</sup>Rn.

#### EVAPORATION/EXTRACTION METHOD

Polymer and solutes

Disperse phase Continuous phase Emulsion Organic solvent removal (and drying)

2
3
4

#### **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



- 2. Slow addition of phases using separation funnel
- 3. Stirring 20h at 25°C
- 4. PSm filtered and cleaned with ethanol and water
- 5. Dried 12h at 40°C



#### **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

#### **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** 



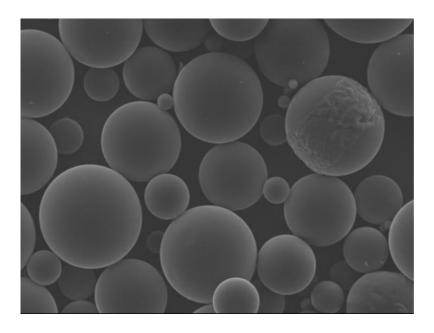
PS/PC 75:25

Agglomerates



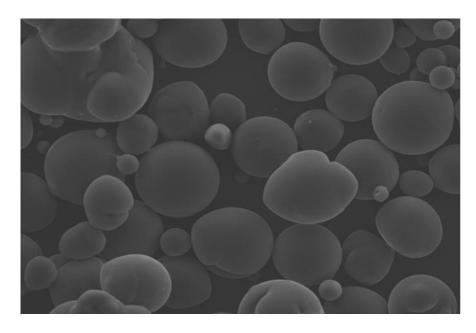
PS/PC 25:75

#### PSm SA (polystyrene)



- Median size 85µm
- Smooth
- Spherical without deformities

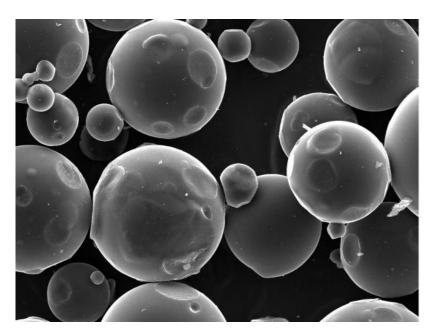
#### PSm MAKD (polycarbonate)



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

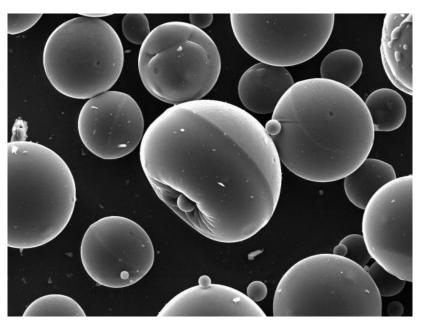
✓ The PC PSm non-spherical form may be caused by the different formation of microdroplets and the process of solidification due to polarity of PC

#### PS/PC (75/25)



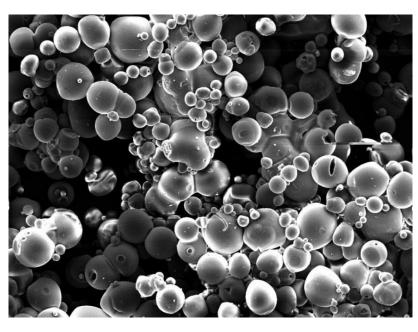
- Agglomerations
- Median size 65µm
- Plains formed in collisions

#### PS/PC (50/50)



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

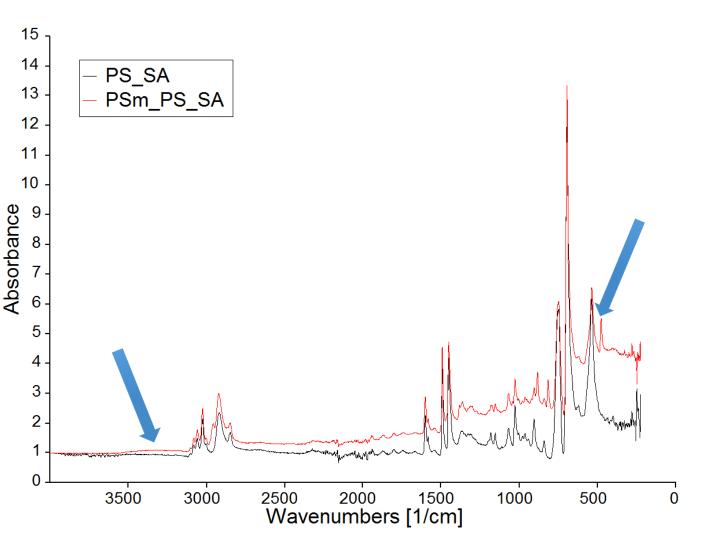
#### **PS/PC (25/75)**



- 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

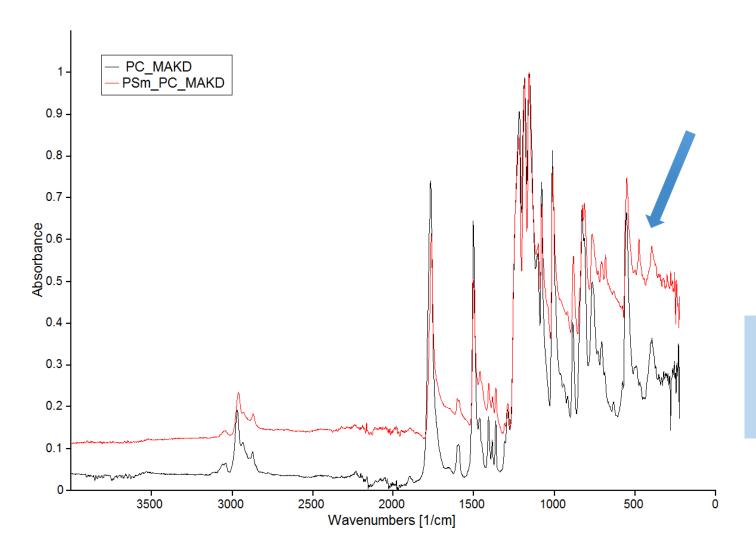
### IR analysis



- PS\_SA (polystyrene)
- PSm\_PS (microspheres of PS)

- ✓ Same peaks (PS is not modified)
- ✓ Encapsulation of solutes
- ✓ Some water and/or PVA?

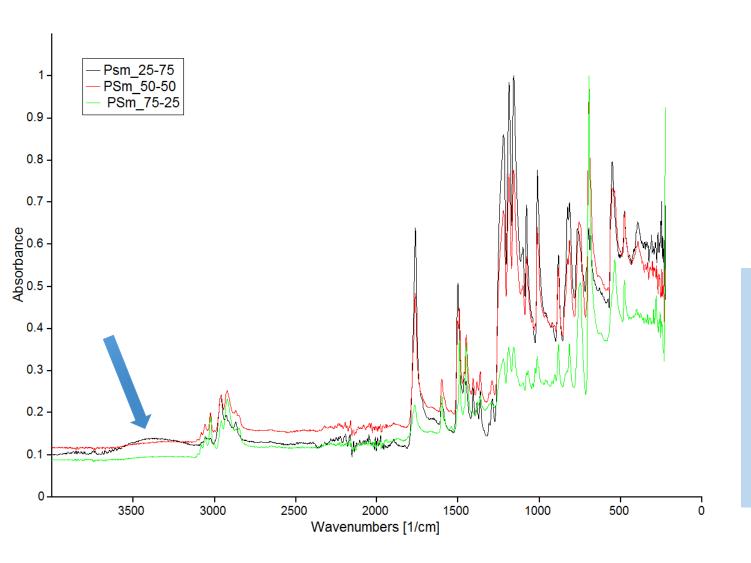
### PSm morphology (IR analysis)



- PC\_MAKD (polycarbonate)
- PSm\_MAKD (microspheres of PC)

- ✓ Same peaks (PC is not modified)
- ✓ Encapsulation of solutes

### PSm morphology (IR analysis)



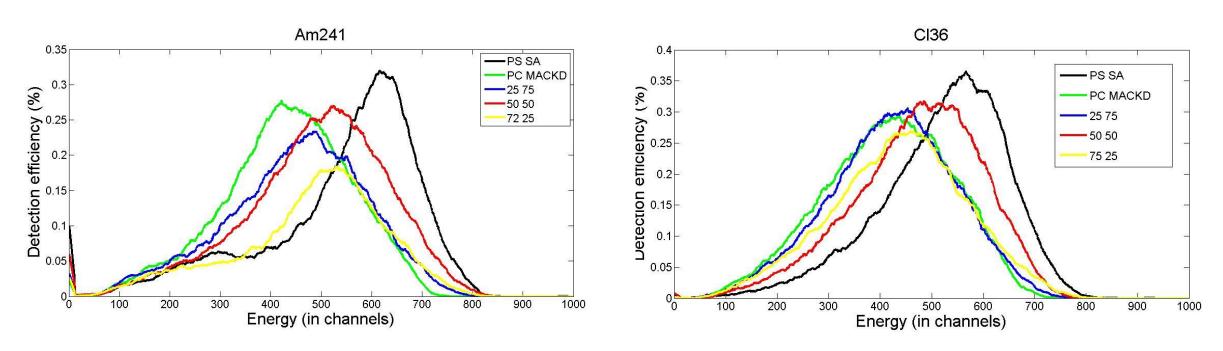
- PSm\_PS\_PC\_75/25
- PSm\_PS\_PC\_50/50
- PSm PS PC 25/75

- ✓ Same peaks (PC and PS are not modified)
- ✓ Encapsulation of solutes
- ✓ Some PVA or water?

#### Quantulus detector

## **Spectrum position and Quenching**





✓ 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. <sup>3</sup> H [%]	0.9(0.1)	0.30(0.03)	0.66(0.03)	0.47(0.03)	0.53(0.02)
Eff. <sup>36</sup> Cl [%]	97(1)	84(3)	96(1)	91(2)	93(1)
Eff. <sup>241</sup> Am [%]	75(1)	53(4)	85(2)	74(1)	77(1)

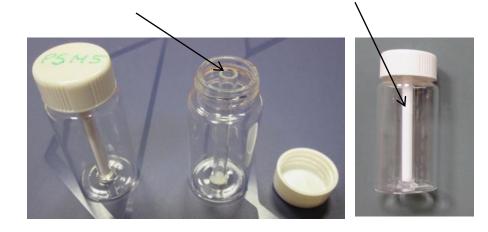
- ✓ Detection efficiency for tritium is low
- ✓ Values are high in all cases except for 75/25 (agglomerations of PSm)
- ✓ Detection efficiency is mainly dependent of the PSm diameter.

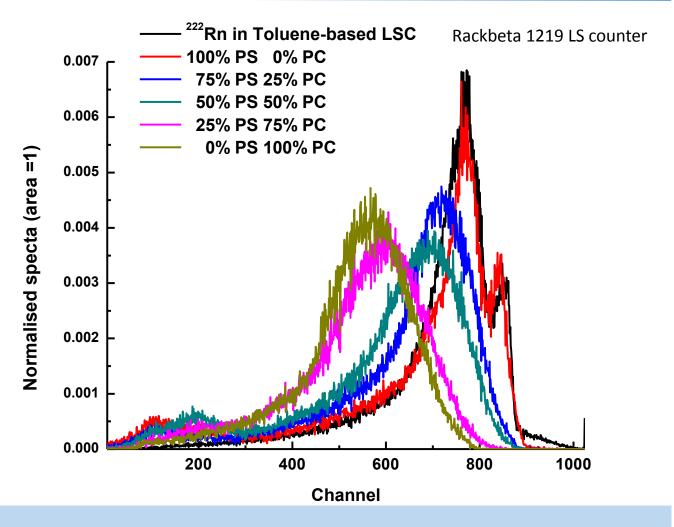
## Radiometric Capabilities

# Spectra of <sup>222</sup>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

# <sup>222</sup>Rn absorption properties

#### Partition coefficients and diffusion lengths

Compare to

PS	m	K		of the raw naterial:
100% PS	0% PC	5.8 (15)	<b>+&gt;</b>	6.62(51)
75% PS	25% PC	6.6 (17)	ľ	
50% PS	50% PC	6.8 (17)		
25% PS	75% PC	5.5 (14)		
0% PS :	100% PC	5.2 (13)	$\leftrightarrow$	26.2(25)

PSm	$L_D$ , $\mu m$ Compare to $L_D$ :
100% PS 0% PC	97.0 (23) 120.8(31)
75% PS 25% PC	69.4 (18)
50% PS 50% PC	49 (14)
25% PS 75% PC	1125 (45) Residual PVA?
0% PS 100% PC	112.9 (46) 52.1(10)

Error introduced by non-spherical PSm?

- ✓ The production of PSm of polycarbobnate changed the radon absorption properties
- $\checkmark$  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 <sup>222</sup>Rn absorption properties of PSm made from 100% polycarbonate differ from the <sup>222</sup>Rn absorption properties of the polycarbonate material.

# Acknowledgment

# This work is supported by the Bulgarian National Science Fund under Contract DFNI T02/13 "POLYRAD"'

Thank you for your attention.

# EVALUATION OF SYNTHESIS CONDITIONS FOR PLASTIC SCINTILLATION FOILS USED TO MEASURE ALPHA- AND BETA-EMITTING RADIONUCLIDES

Merín, R.1; Tarancón, A.1,3; Mitev, K.2; Georgiev, S.2; Dutsov, Ch.2; Bagán, H.1; García, J.F.1

<sup>&</sup>lt;sup>1</sup>Department of Chemical Engineering and Analytical Chemistry, University of Barcelona, Marti i Franqués, 1-11, ES-08028, Barcelona, Spain.

<sup>&</sup>lt;sup>2</sup>Faculty of Physics, Sofia University "St. Kliment Ohridski", 1164 Sofia, Bulgaria.

<sup>&</sup>lt;sup>3</sup>Serra-Húnter Program, Generalitat de Catalunya, Barcelona Spain.

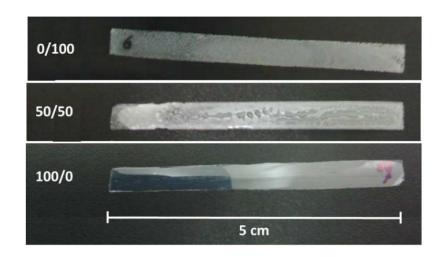
# EVALUATION OF SYNTHESIS CONDITIONS FOR PLASTIC SCINTILLATION FOILS USED TO MEASURE ALPHA- AND BETA-EMITTING RADIONUCLIDES

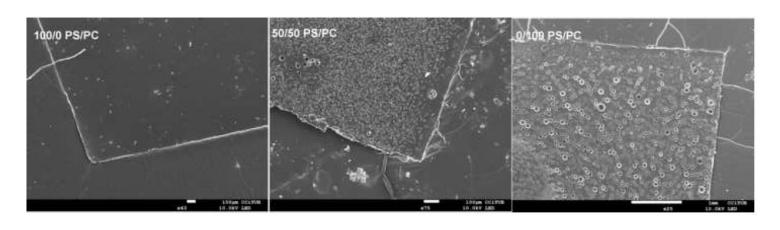
Merín, R.1; Tarancón, A.1,3; Mitev, K.2; Georgiev, S.2; Dutsov, Ch.2; Bagán, H.1; García, J.F.1

<sup>&</sup>lt;sup>1</sup>Department of Chemical Engineering and Analytical Chemistry, University of Barcelona, Marti i Franqués, 1-11, ES-08028, Barcelona, Spain.

<sup>&</sup>lt;sup>2</sup>Faculty of Physics, Sofia University "St. Kliment Ohridski", 1164 Sofia, Bulgaria.

<sup>&</sup>lt;sup>3</sup>Serra-Húnter Program, Generalitat de Catalunya, Barcelona Spain.





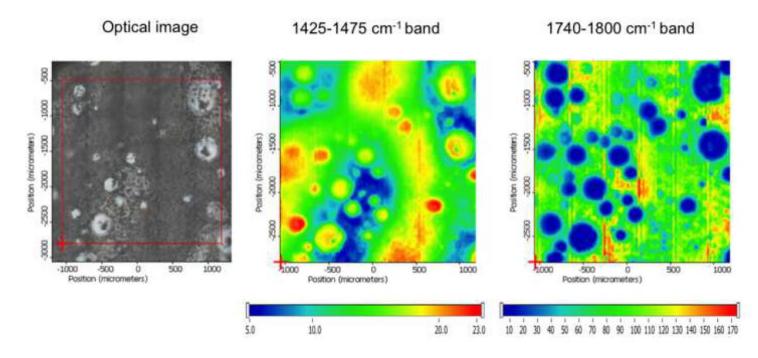


Table 7: Partition coefficients (K) for <sup>222</sup> Rn form air of PS foils with different polystyrene/polycarbonate composition (%). The values are for 21°C.				
Partition coefficient (solubility)				
11.7(28)				
7.3(17)				
4.1(13)				
5.0(11)				
14.9(30)				

Table 8: Diffusion lengths ( $L_D$ ) of <sup>222</sup> Rn in 100% polystyrene or 100% polycarbonate foils. The values are for 21°C.					
PS composition: polysterene/polycarbonate concentration (%)	Diffusion length $(L_D)$ , $\mu$ m				
0/100	259(61)				
100/0	51(17)				

Applied Radiation and Isotopes 109 (2016) 270-275



Contents lists available at ScienceDirect

#### Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso



Determination of <sup>222</sup>Rn absorption properties of polycarbonate foils by liquid scintillation counting. Application to <sup>222</sup>Rn measurements



K. Mitev <sup>a,\*</sup>, P. Cassette <sup>b</sup>, S. Georgiev <sup>a</sup>, I. Dimitrova <sup>a</sup>, B. Sabot <sup>b</sup>, T. Boshkova <sup>a</sup>, I. Tartès <sup>b</sup>, D. Pressyanov a

Table 1 LD and K for 222Rn from air for Makrofol N and DE foils.

Type of foil	Diffusion length, $L_D$ ( $\mu m$ )	Partition coefficient from air, K
Makrofol N	38.9 ± 1.3	$112 \pm 12$
Makrofol DE	52.1 ± 1.0	$26.2 \pm 2.5$

a The values are for 20 °C.

a Faculty of Physics, Sofia University "St. Kliment Ohridski", 1164 Sofia, Bulgaria

b CEA, LIST, Laboratoire National Henri Becquerel, LNE-LNHB, Gif-sur-Yvette F-91191 France

#### **Final remarks:**

- It will probably be beneficial if we gain more information about the production way of different polycarbonate- based products: pellets, CDs, DVDs, foils or even powder, since this information may be related to the noble gas absorption properties.
- ➤It will be perhaps useful to test same material (e.g. Makrofol DE) produced in different ways: extrusion, meltcasting, extraction-evaporation, etc., because these might enhance or suppres noble gas absorption properties.