Study of the Partition Coefficient and the Diffusion Length of Radon in Polymers at Different Temperatures: Experimental Approach and Results

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SUBG

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Polymer membranes

- Radon/Thoron discrimination in lots of passive detectors
- Radon mitigation by water-proofing membranes

Both count on “diffusion delay” $L_D = \sqrt{\frac{D}{\lambda}} \implies L_D \sim \sqrt{T_{1/2}}$

… and D , resp. L_D are temperature dependent

Polymer samplers

- Methods for radon measurements are developed, based on the high radon absorption ability in polymers such as Makrofol DE and N

- The absorption could be described by the partition coefficient K and the diffusion length L_D and they both depend on the temperature

Model of RNG transport in polymers

- At the border polymer/ambient media the RNG concentration ratio is:

$$K = \frac{C_{in}}{C_{out}}$$

- Once in the polymer, the radioactive noble gas (RNG) diffuses and decays:

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - \lambda c$$

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Sorption and desorption of radioactive noble gases in polycarbonates

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Model of RNG transport in polymers

- And the absorbed activity in thin plate is:

$$A(t_s, t_d) = \frac{8\lambda L_D^2 VKC_A}{L^2} \sum_{k=0}^{\infty} \frac{e^{-\lambda t_s} - e^{-\lambda_k t_s}}{\lambda_k - \lambda} e^{-\lambda_k t_d},$$

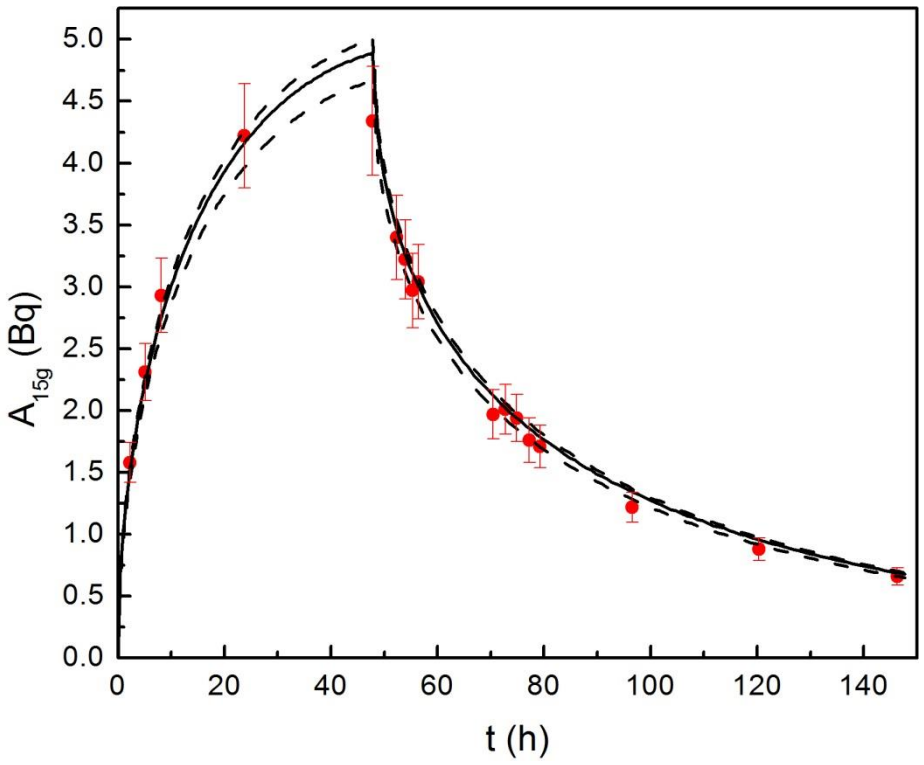
$$\lambda_k = \lambda \left(1 + \left(\frac{(2k+1)\pi L_D}{L} \right)^2 \right),$$

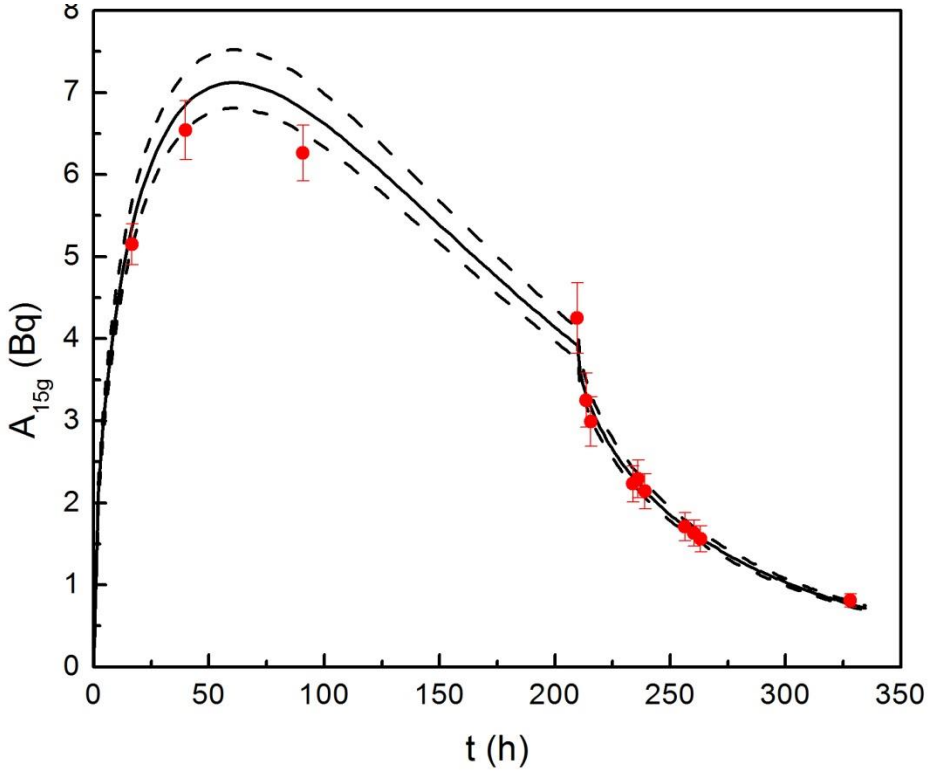
λ – RNG decay const.

L, V – plate thickness and volume

C_A – ambient RNG concentration at t_s=0

t_s , t_d – sorption and desorption time



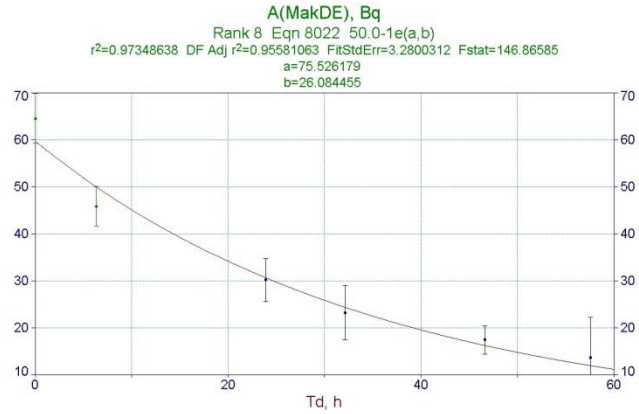


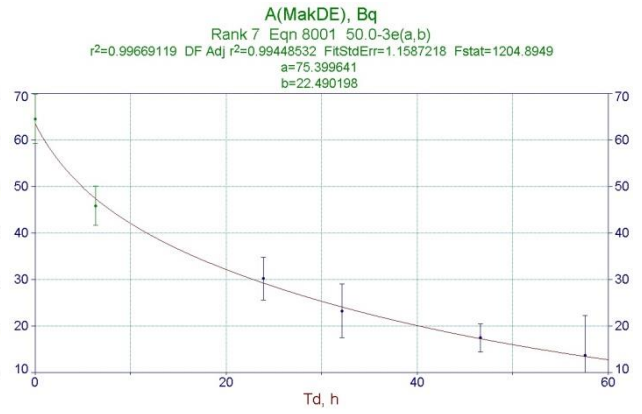
Method for estimation of K and L_D

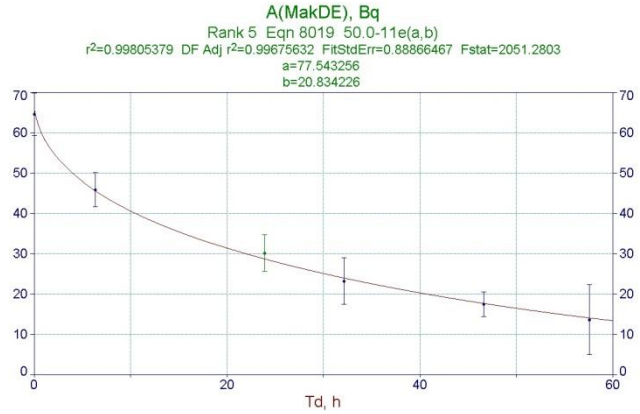
- The absorbed-activity equation could be rewritten:

$$A(t_d; K, L_D) = 8VC_AK \sum_{n=1}^{\infty} \frac{e^{-\lambda t_s} - e^{-\lambda\left(1+(n\pi)^2\left(\frac{L_D}{L}\right)^2\right)t_s}}{(n\pi)^2} e^{-\lambda\left(1+(n\pi)^2\left(\frac{L_D}{L}\right)^2\right)t_d}$$

$n = (2k+1)$ – an odd number







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DETERMINATION OF THE DIFFUSION COEFFICIENT AND SOLUBILITY OF RADON IN PLASTICS

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Desorption follow-up: challenges

- Temperature have to be constant
  - During both sorption and desorption
  - Including the activity measurement

- Desorption could be fast – depending on L_D/L it could take a few minutes
  - Fast and precise timing

- Secular equilibrium between Radon/SLP is needed
  - Radon is measured by its short-lived progeny (SLP)
  - So the fast desorption must be “stopped” for the measurement duration (or somehow accounted…)

Desorption follow-up: challenges

- LSC with toluene cocktail is the best decision for Makrofol N – its fully dissolved in toluene

- …but doesn't work for the other polymers

  – Makrofol DE is only partially dissolved in toluene
  – PP, HDPE, LDPE not dissolved at all in toluene and other organic solvents: Gasoline, Bensol(Benzene), 1,2-Dichloroethane (ethylene dichloride)
  – Activity desorbs in the cocktail and the cocktail (or at least toluene) is absorbed in the polymers

  => ***Complicate change in the Counting efficiency***

Desorption follow-up: challenges

- Cherenkov counting in water:

  + Allows precise timing

  + Almost fully stops the activity loss during the measurement and allows a posteriori correction

  + No temperature control during the measurement

  + One point of the desorption could be measured more than once

  – Radon desorbs in the water until equilibrium of radon concentration in the two media is reached

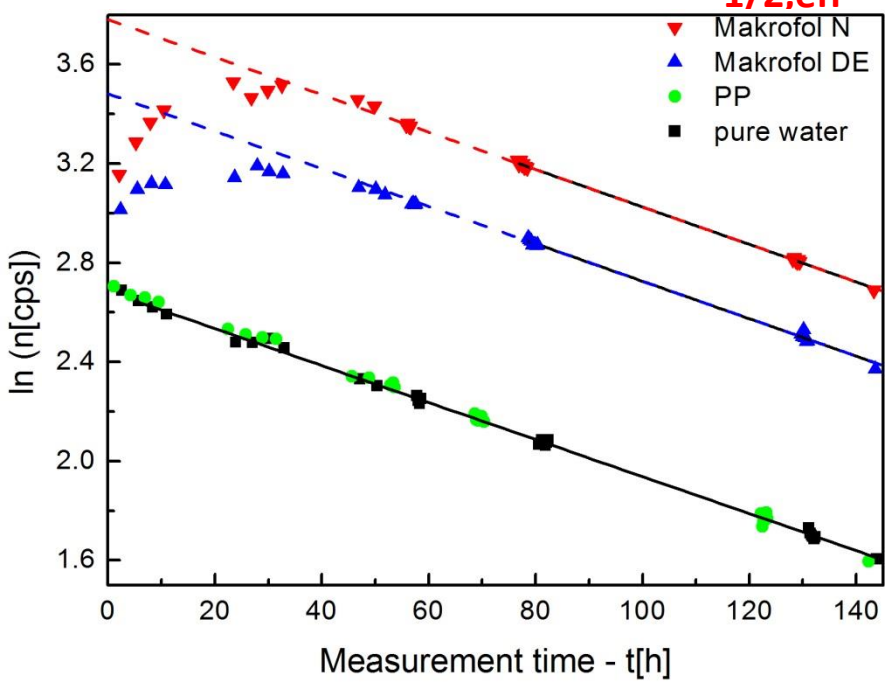
  $\Rightarrow$ Change of the efficiency during that process

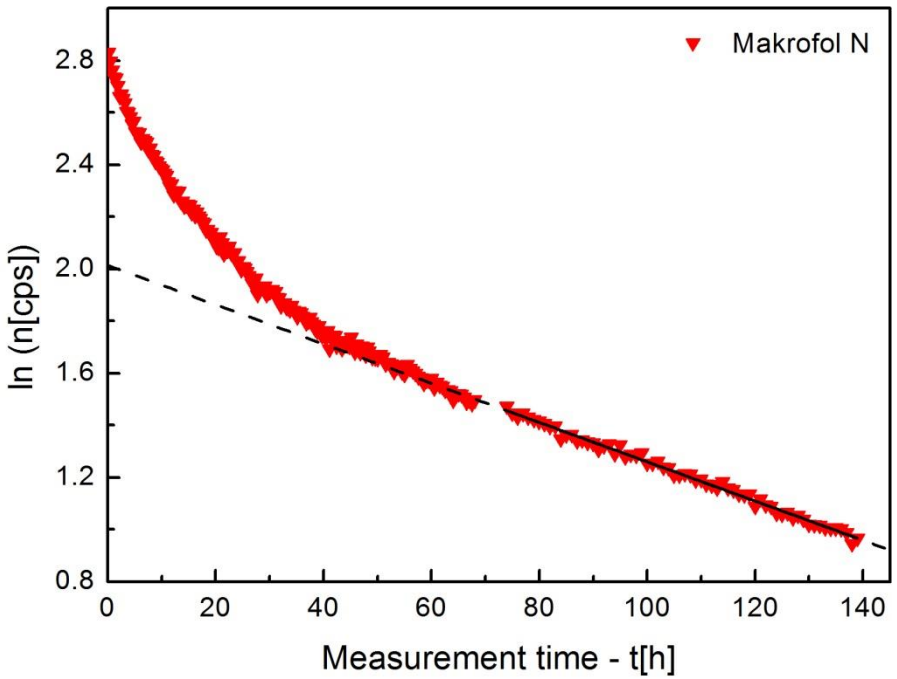
  – Each point of desorption is a different foil

Counting efficiencies ε_c

- Two experiments for ε_c estimation:
  - Unexposed foils in high-activity water for better counting statistics
  - Exposed Makrofol N in distilled water – to compare with the previous experiment

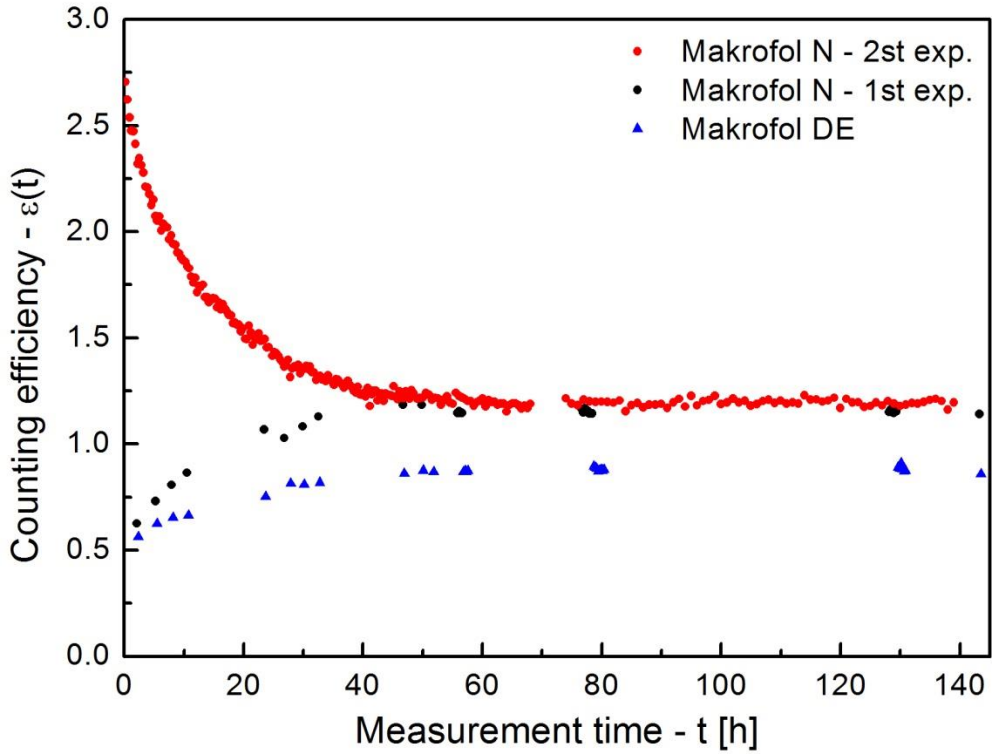
$T_{1/2,eff}$=89.5(9)h





Counting efficiencies ε_c

- Samples are followed at the RackBeta LS-counter and measured several times at HPGe
  - No time dependence of ε_c for PP, LDPE, HDPE
  - After 60-70h ε_c=*const* for Makrofols

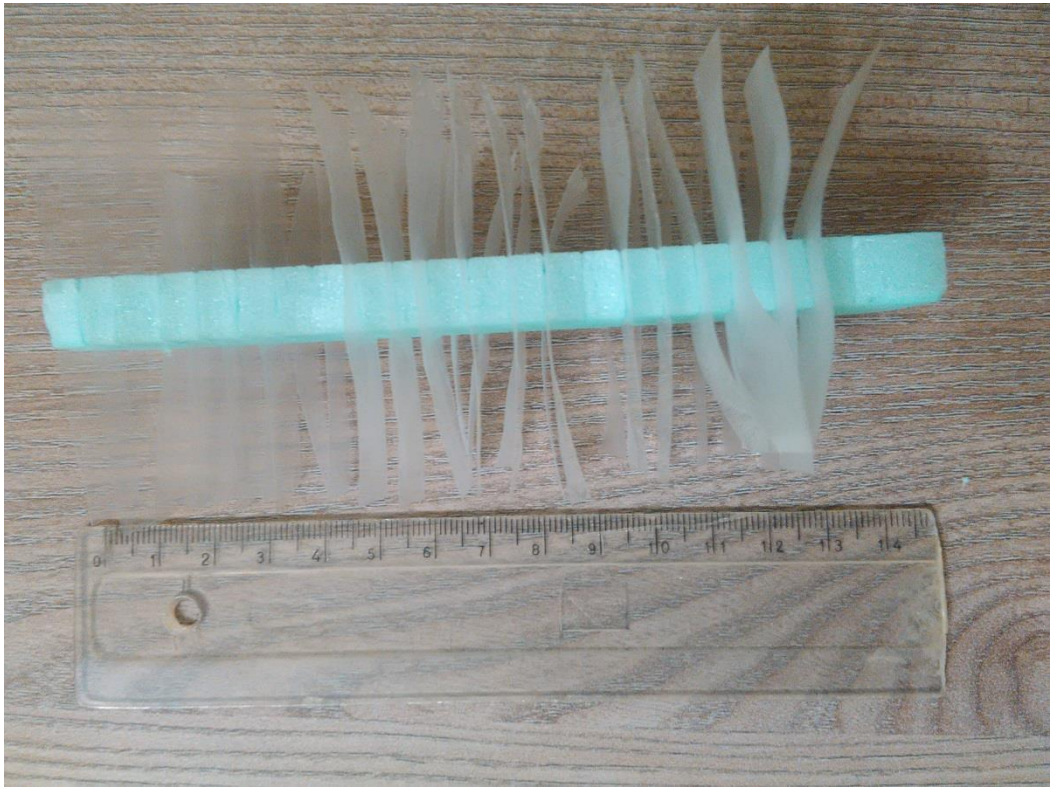


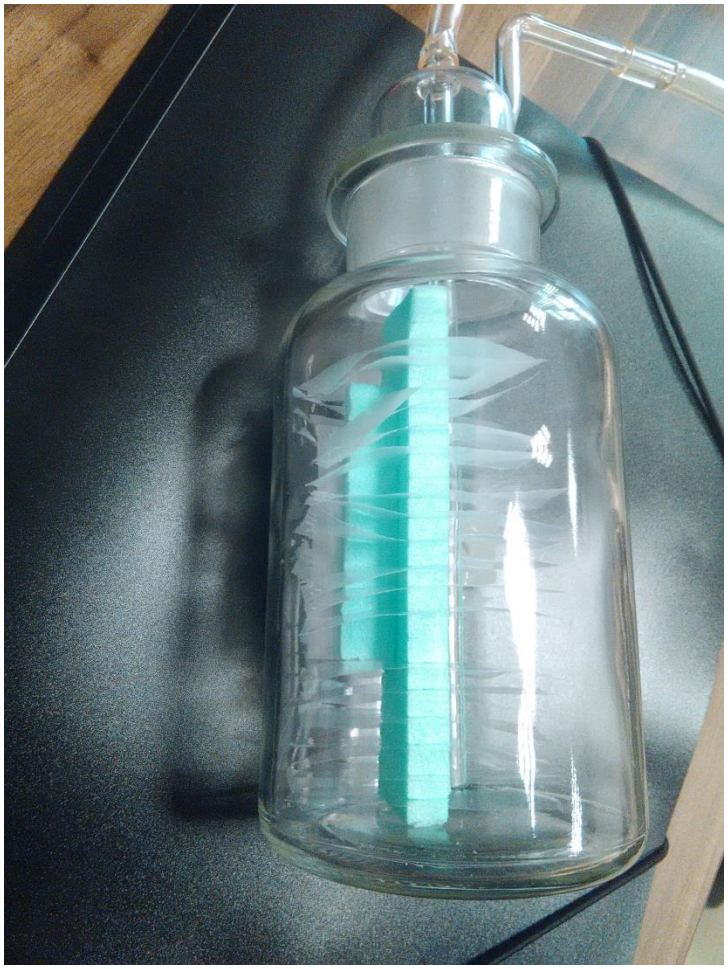
No	Sample	Counting efficiency
1	PP in water	0.380(12)
2	LDPE in water	0.371(12)
3	LDPE-A in water	0.400(14)
4	HDPE in water	0.407(13)
5	Makrofol N in water	1.168(36)
6	Makrofol DE in water	0.883(29)
7	pure water	0.376(12)
8	Makrofol N in LSC	4.946(29)

K, L_D experiments: Exposure setup

- Six foils 5.6cm x 1.6cm of each material:

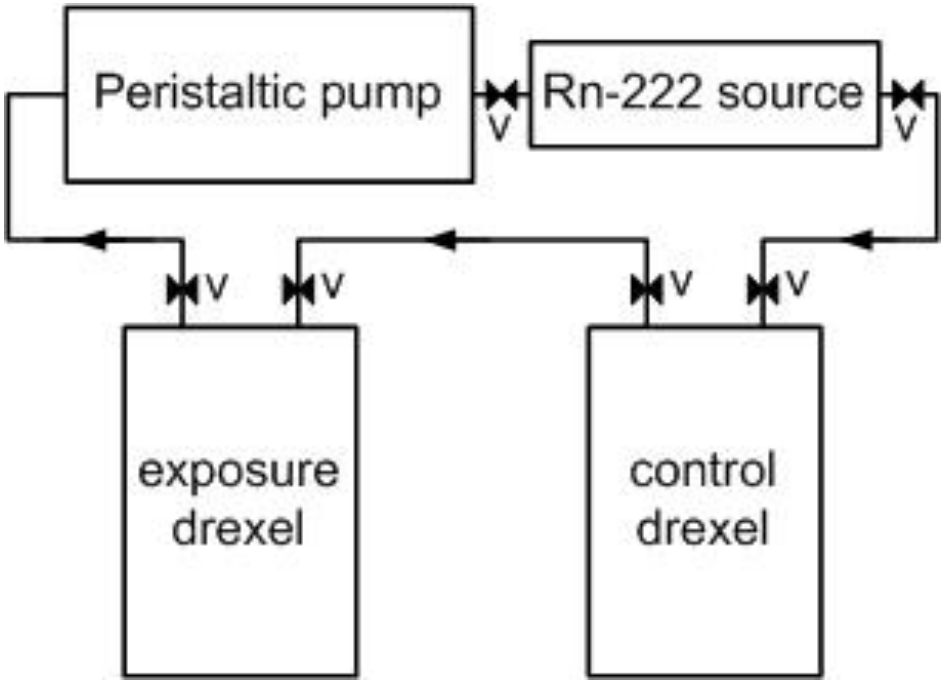
PP, LDPE, LDPE-A, HDPE, Makrofol DE, Makrofol N



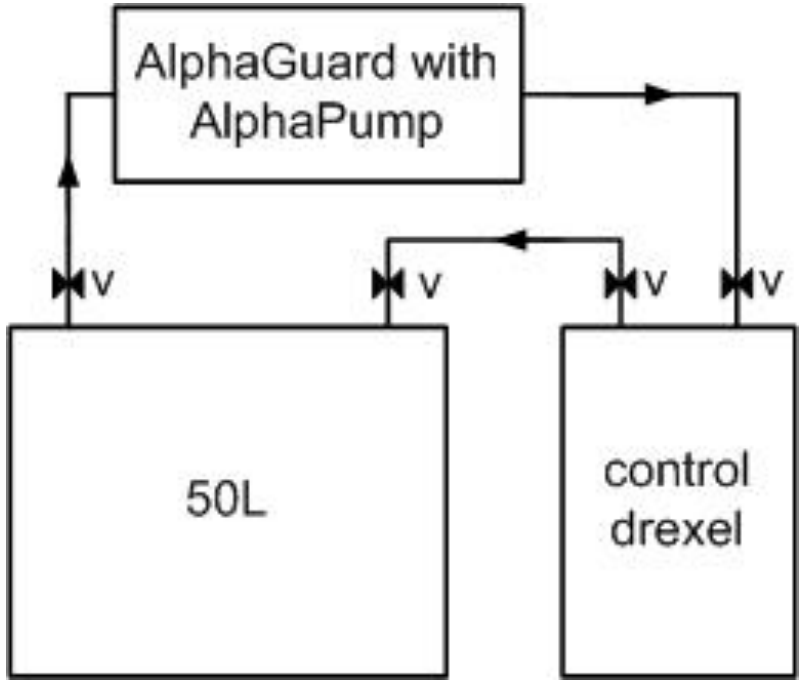


K, L_D experiments: Exposure setup

- Three experiments with very high activity in small volume at 5°C, 21°C and 31°C
  - High C_A to ensure better counting statistics
  - and longer desorption follow-up



Rn is promptly introduced in the system in the beginning and the system is disconnected



The activity from the control drexel is diluted and measured, in order to estimate the exposure C_A

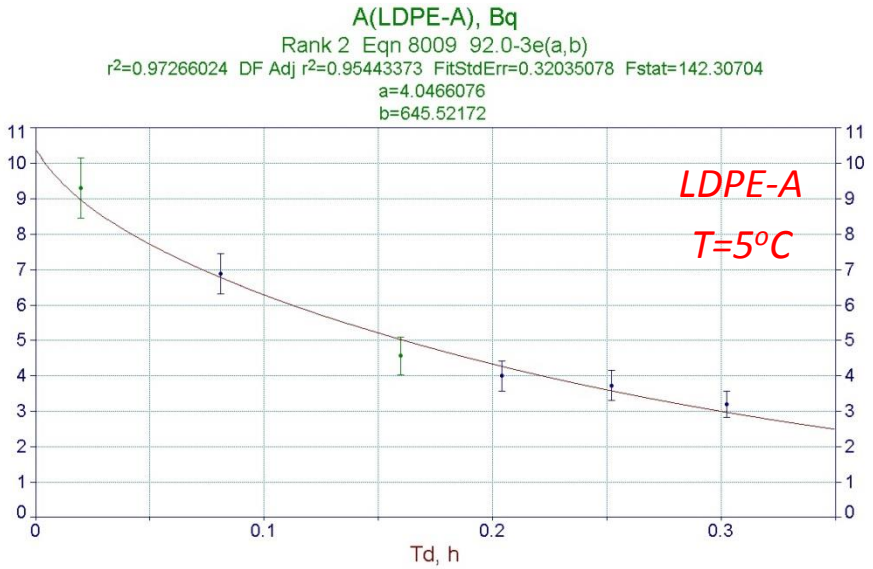
K, L_D experiments: Exposure setup

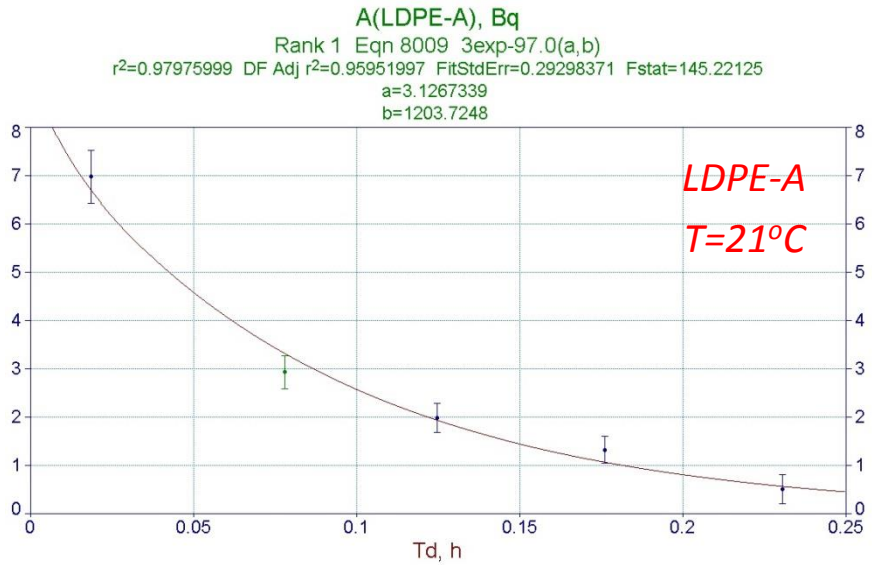
- During the exposure, the exposure drexel was placed in the 50L box,and the 50L box was placed in thermostate for temperature control
- The activity concentration in the 50L box was measured in order to check for activity leakage
  - The leakage was fond to be less than 1% in all experiments
- After the exposure the foils were kept in the thermostate, in order to desorb at the same temperature

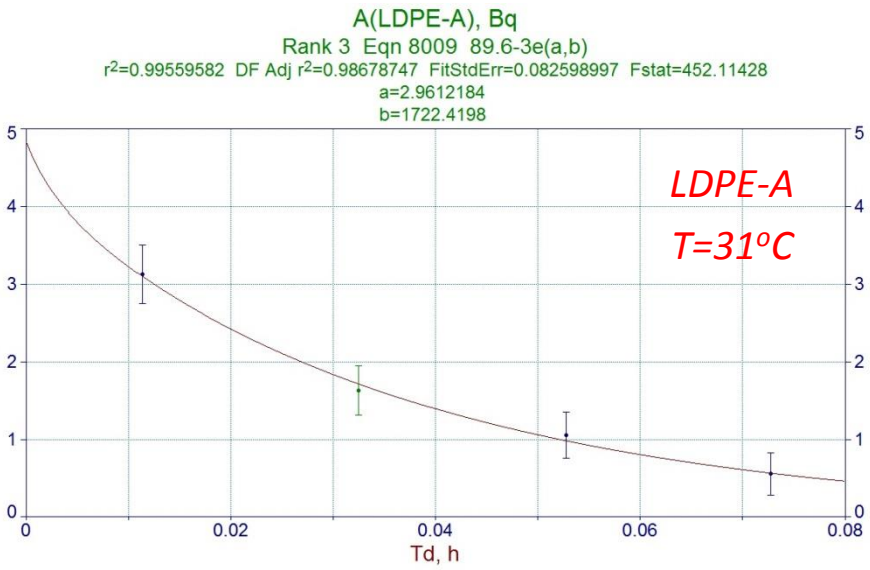
K, L_D experiments: Exposure setup

- One experiment in the 50L referent volume of the AlphaGuard at 10^{o}C
  - Due to much lower C_A, only Makrofol foils were used
  - The foils were stuck in the holder and placed directly in the 50L volume
  - C_A was measure directly
  - 6 Makrofol DE and 12 Makrofol N foils were used
  - 6 Makrofol N were measured in toluene and the other 6 – in water

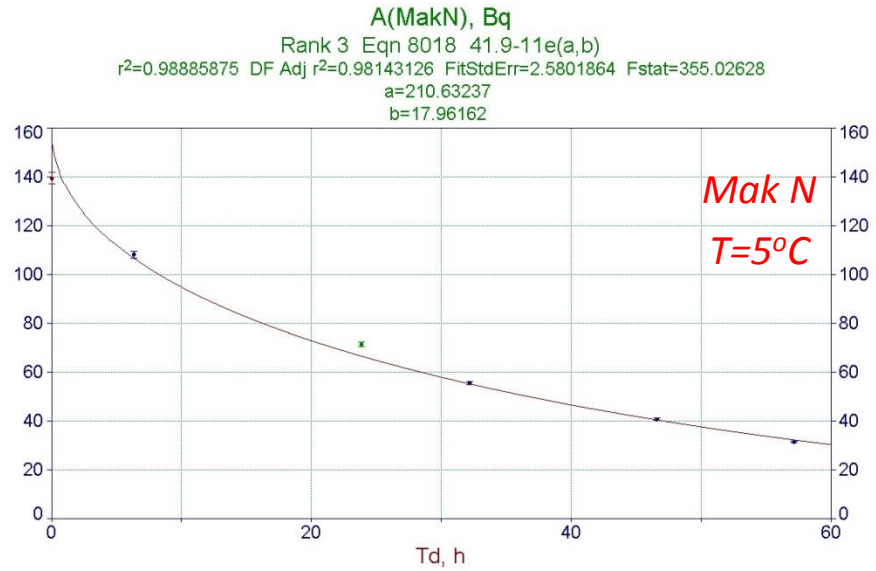
K, L_D experiments: Results

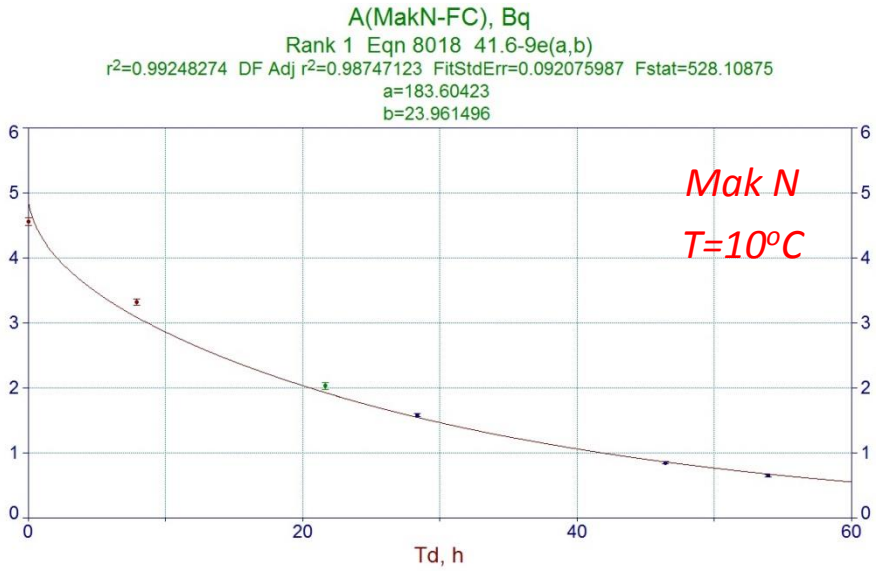


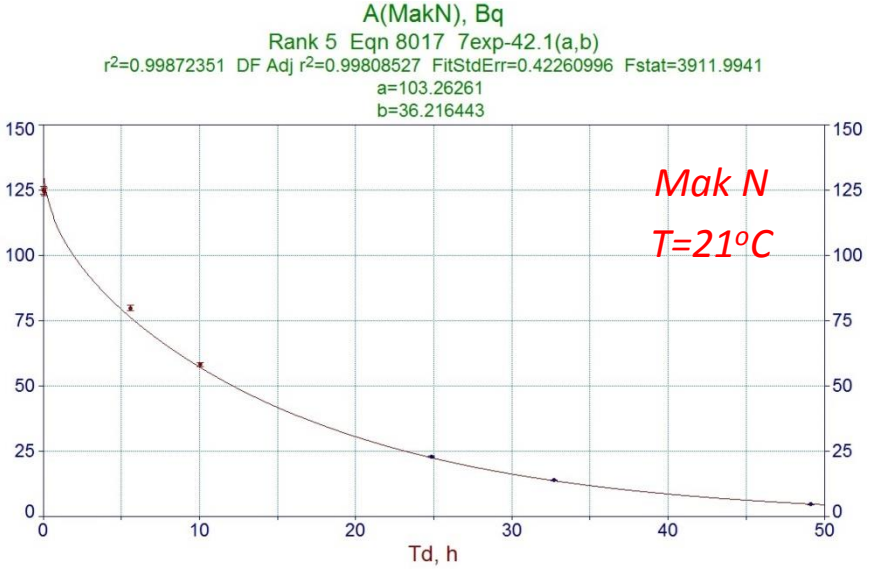


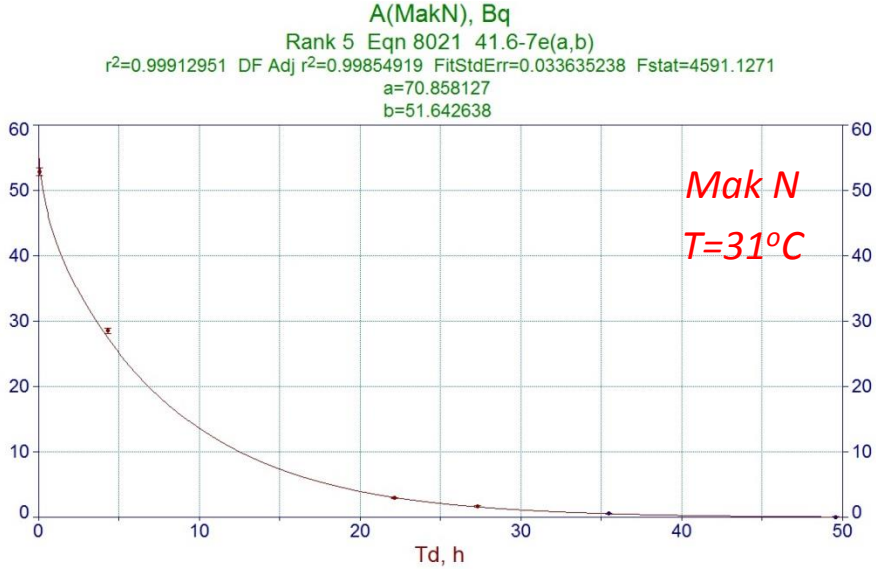


K, L_D experiments: Results





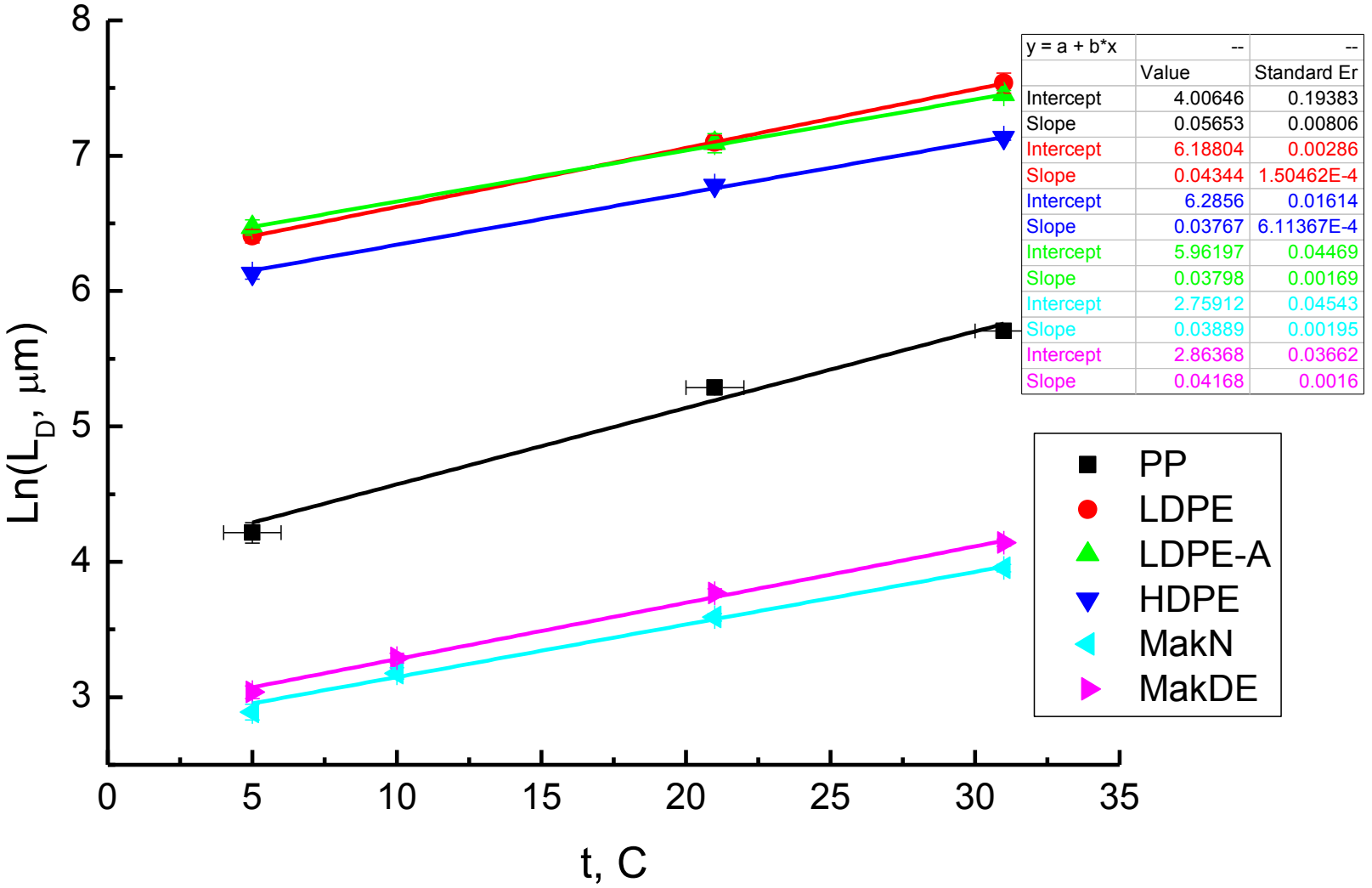




K, L_D experiments: Results

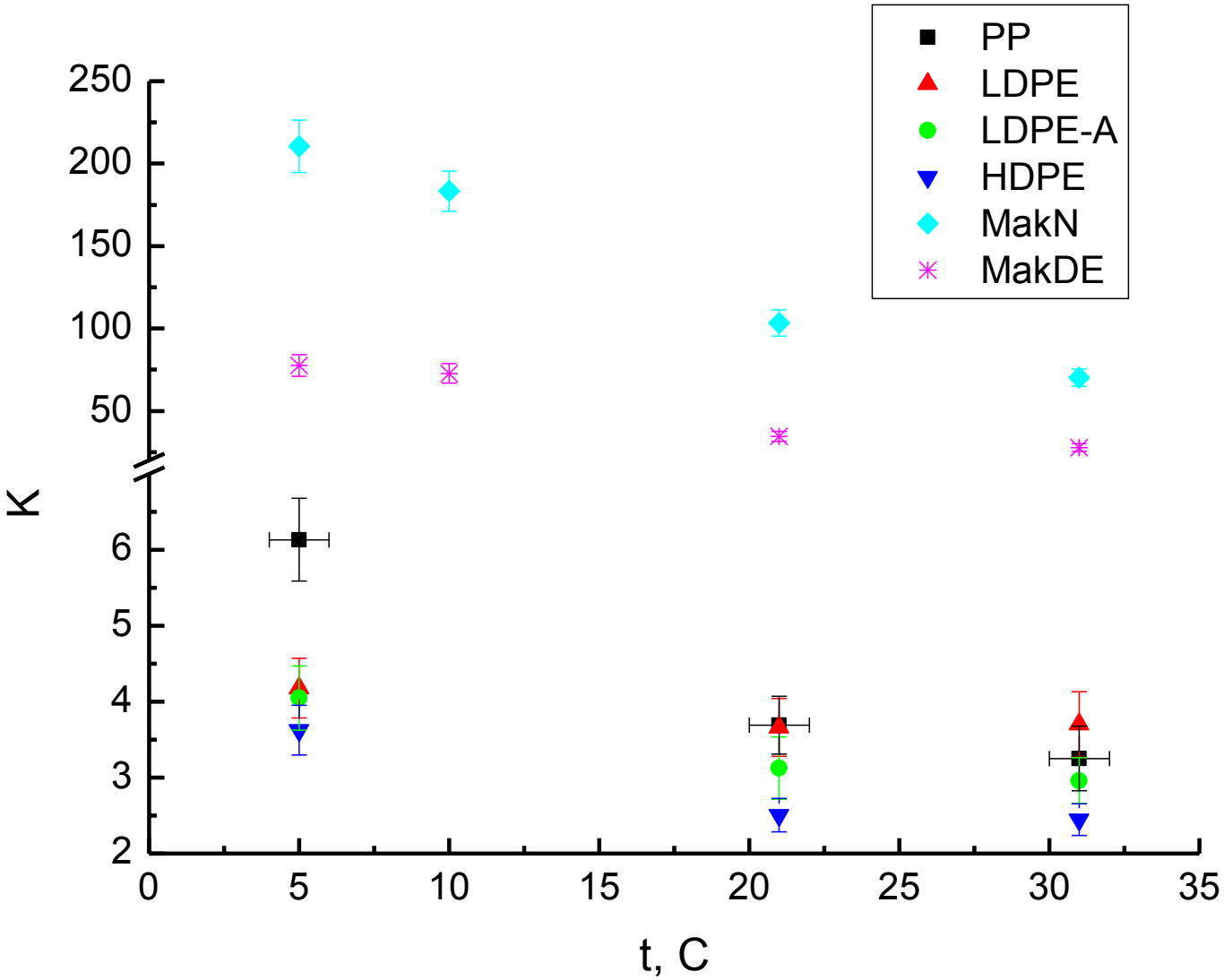
	PP	LDPE	LDPE-A	HDPE	Mak DE	Mak N	MakDE*	
T°C	Partition coefficient K							T°C
5(1)	6.13(55)	4.18(39)	4.05(42)	3.63(33)	77.5(67)	211(16)	21.5(43)	5
10(1)					72.8(58)	183(12)	24.3	10
21(1)	3.69(38)	3.66(38)	3.13(41)	2.51(22)	34.6(30)	103.3(79)	26.4(25)	19.5
31(1)	3.25(43)	3.70(43)	2.96(30)	2.44(21)	27.8(24)	70.2(51)	22.9	31
~20		2.17(14)		2.21(13)	27.6(16)	112(12)		
		2.40(22)						
ToC	Duffusion length L_D, um							T°C
5(1)	67.6(51)	605(30)	646(36)	460(19)	20.8(10)	18.0(10)	42.2(16)	5
10(1)					26.8(10)	23.9(10)	42.8	10
21(1)	198(10)	1210(64)	1204(85)	880(22)	43.3(13)	36.2(10)	51.7(8)	19.5
31(1)	300(15)	1880(140)	1722(54)	1252(23)	62.9(16)	52.1(15)	75.5	31
~20		1463(33)		721(9)	50.8(10)	38.9(13)		
		1437(94)						
ToC	Diffusion coefficient D, 10^{-14} m^2/s							
5(1)	0.96(14)	76.9(77)	87.4(97)	44.3(37)	0.0911(84)	0.0677(79)		
10(1)					0.151(11)	0.120(10)		
21(1)	8.20(85)	307(33)	304(43)	162(8)	0.394(25)	0.275(15)		
31(1)	18.9(19)	739(111)	623(39)	329(12)	0.831(43)	0.570(32)		
~20		448(10)		109(2)	0.540(12)	0.318(11)		
		432(28)						
ToC	Permeability P, 10^{-13} m^2/s							
5(1)	0.59(10)	32.1(44)	35.4(54)	16.1(20)	0.706(89)	1.43(20)		
10(1)					1.10(12)	2.20(24)		
21(1)	3.03(44)	113(17)	95.1(18)	40.7(41)	1.36(15)	2.84(27)		
31(1)	6.1(10)	273(52)	184(22)	80.4(75)	2.31(23)	4.00(37)		
~20		97.2(66)		24.0(15)	1.49(9)	3.56(40)		
		104(9)						
	Pressyanov et al, Rad. Prot. Dosim. 145(2-3) (2011) 123-126							
	Mitev et al, Appl. Rad. Isot.109 (2016) 270–275							
	Pressyanov, HealthPhys 2009							

K, L_D experiments: Results

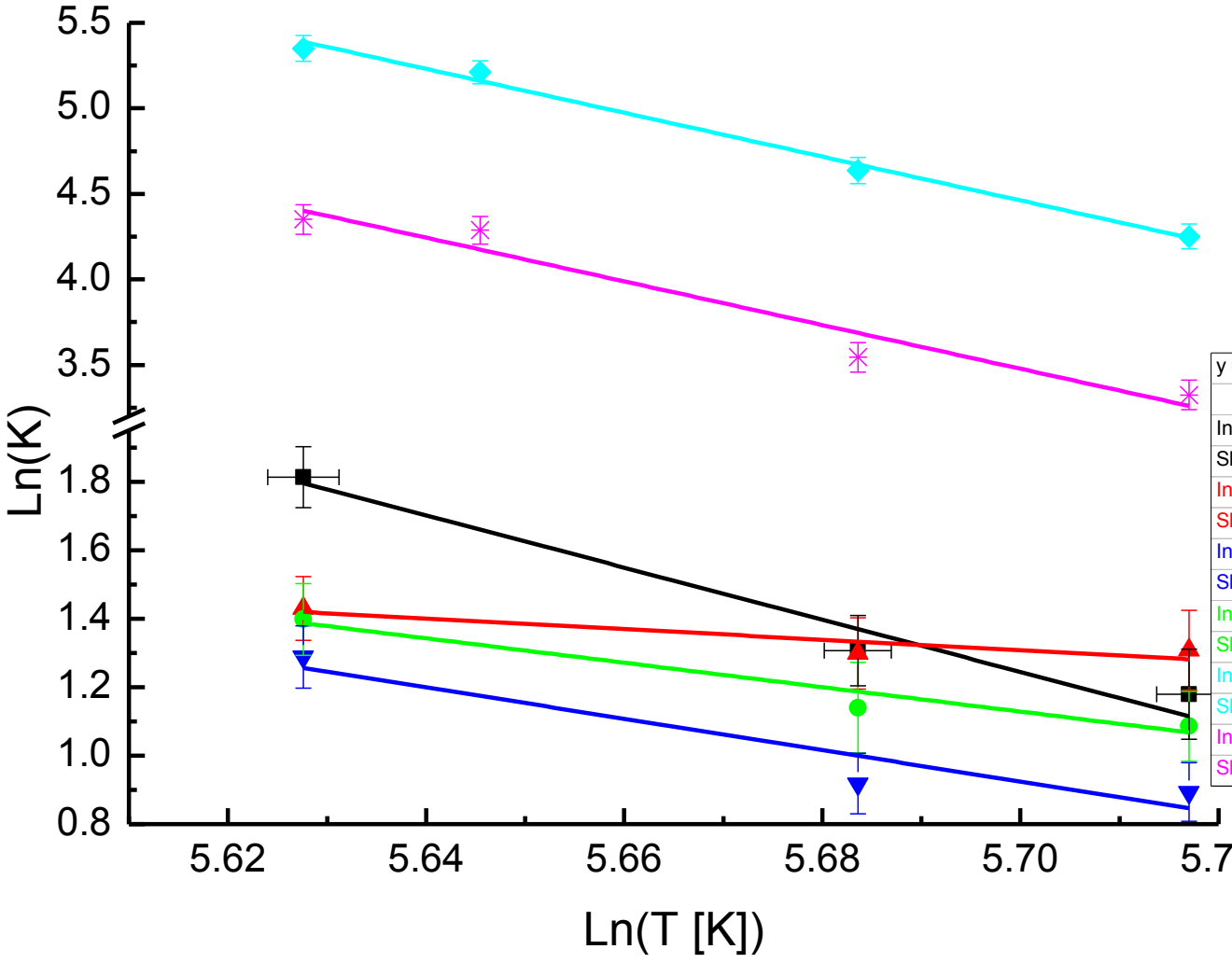


y = a + b*x	--	--
	Value	Standard Er
Intercept	4.00646	0.19383
Slope	0.05653	0.00806
Intercept	6.18804	0.00286
Slope	0.04344	1.50462E-4
Intercept	6.2856	0.01614
Slope	0.03767	6.11367E-4
Intercept	5.96197	0.04469
Slope	0.03798	0.00169
Intercept	2.75912	0.04543
Slope	0.03889	0.00195
Intercept	2.86368	0.03662
Slope	0.04168	0.0016

K, L_D experiments: Results



K, L_D experiments: Results



y = a + b*x		
	Value	Standard Error
Intercept	44.66553	7.60899
Slope	-7.61774	1.34309
Intercept	10.12592	3.84883
Slope	-1.54708	0.67885
Intercept	21.46541	3.79918
Slope	-3.56787	0.66936
Intercept	27.07127	8.87171
Slope	-4.58721	1.5625
Intercept	77.52242	4.36118
Slope	-12.81769	0.76948
Intercept	76.28292	11.77789
Slope	-12.77287	2.07811

Thank you!