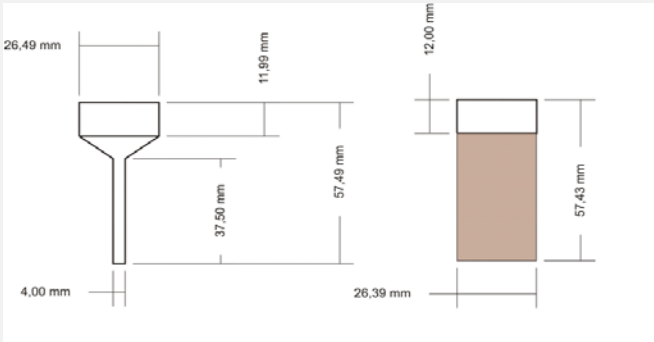
The comparison of scintillation properties of YAP:Ce, YAG:Ce and ZnO:Ga powders as a potential substitution of LSC cocktail

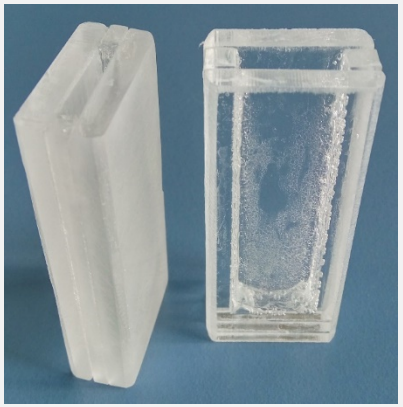
Introduction

- **Why?**
  - LSC cocktail is liquid organic chemical → problems with transportation, storage, handling, disposal → problems of field lab and to the future.
- **Need for safe and inert scintillation means.**
  - Solid scintillation is the ultimate step in LS technique evolution solving all above mentioned problems.
- **Associated problems:**
  - Long-term stability of powder, radionuclide distribution throughout the volume, shape of the spectra

Research origin

- Use of YAP:Ce powder of micron grain sizes in connection with acid environment
  - maximum efficiency is approximately two third of efficiency of scintillation cocktail
  - quenching process is almost negligible → direct sample measurement after different treatment methods (MW digestion, acid leaching) – without the need of the quenching curve.
  - Excellent thermal stability, very small afterglow, repeatability usage,...
  - Strong influence of geometry measurement, spectrum deterioration
  - New type of the vials



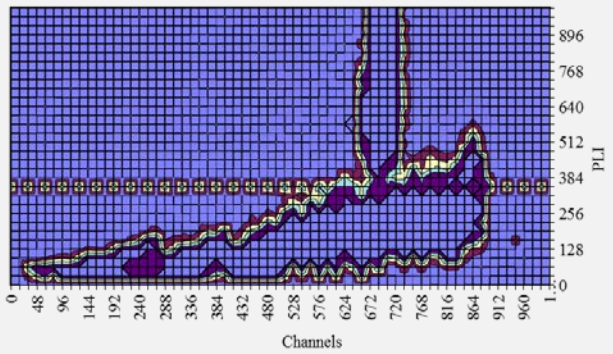


Results and outcomes

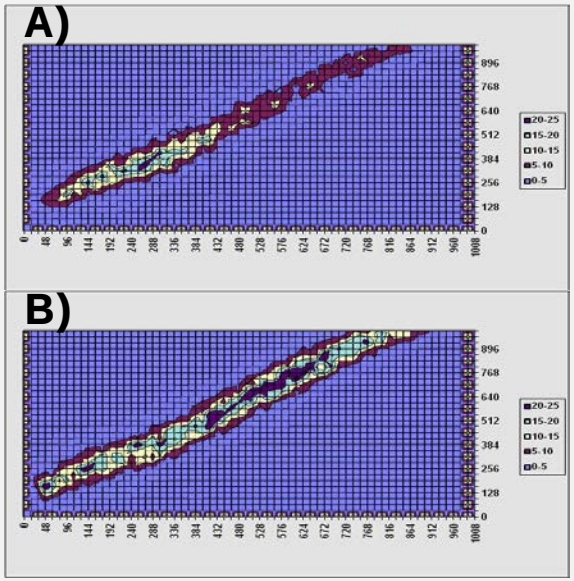
Physical properties		Surface type	Rotation	^{241}Am		Δ from 80 ± 3 %		^{90}Sr		Δ from 73 ± 2 %	
				Eff[%]	σ[%]	Eff[%]	σ[%]	Eff[%]	σ[%]	Eff[%]	σ[%]
Vial spacing [mm] / mass of powder [g]	2,9 - 3,5	Shiny	PMT-Flat	85.7	2.5	**5.7**	**3.9**	66.8	4.0	**-6.2**	**4.5**
			PMT-Edge	83.1	1.6	**3.1**	**3.4**	66.2	2.8	**-6.8**	**3.4**
		Opaque	PMT-Flat	84.7	2.1	**4.7**	**3.6**	69.0	2.9	**-4.0**	**3.5**
			PMT-Edge	83.7	1.3	**3.7**	**3.3**	66.2	1.9	**-6.8**	**2.8**

Comparison of detection efficiencies of proposed vials with the optimal values stated in section “Detection efficiency and quenching”. Δ means difference between detection efficiency of proposed vials and detection efficiency of optimal value.

2D spectrum of ^{241}Am and ^{90}Sr using cocktail Aqualight



2D spectrum of ^{241}Am (A) and ^{90}Sr (B) using YAP:Ce powder of grain size of 70-90 μm and 250-315 μm respectively



theorem

Many of the radiochemical species of interest behave the same as trace metals. The preservation of water samples is easily achieved by acidification which prevents metallic species from depositing on the walls of the container.

Support the sorption on the surface of the scintillation powder → use alkaline environment.

Aim of the research

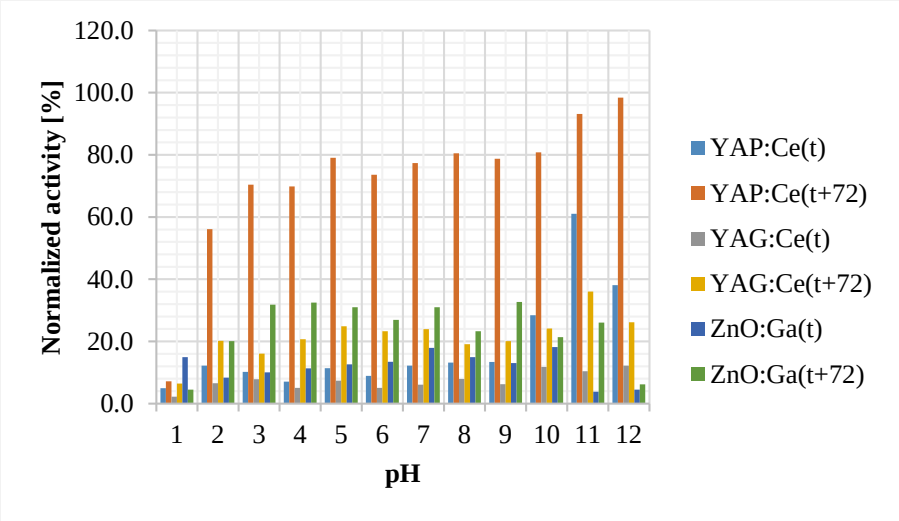
- Response of YAP:Ce (0-30 µm), YAG:Ce (30 µm and 30 nm) and ZnO:Ga (5 µm) to alpha and beta emitters.

Primary research interest:

- pH dependency
- OH^- dependency
- The amount of the scintillator
- Temperature influence
- Afterglow
- Redox agents influence
- Shape of the spectra
- Linear dependence of the activity

pH dependency

- Validation of sorption theory of radionuclides onto a surface under high pH, as a powerful tool and basic assumption of this solid scintillation type technique.



pH dependency of ^{232}U on scintillation yield expressed by normalized activity. The measurement was performed immediately after shaking (t) and 72 hours after shaking (t+72). The yield was normalized to "true" activity added to the vial.

Scintillation yield [%], pH 12						
Nuclide	YAP:Ce		YAG:Ce		ZnO:Ga	
	t	t+72 h	t	t+72 h	t	t+72 h
^{241}Am	31.2	83.6	1.2	12.8	3.9	17.5
^{239}Pu	16.4	74.0	9.3	27.8	3.9	12.1
^{232}U	38.0	98.4	12.3	26.2	4.5	6.2
^{90}Sr	11.9	22.3	17.3	24.0	3.1	7.6

Normalized activities of investigated radionuclides and scintillation powders at pH 12. The measurement was performed immediately after shaking (t) and 72 hours after shaking (t+72).

OH^- dependency

Dependency of scintillation yield on different concentration of solution of sodium hydroxide after 38 hours. The italic type of the font is used for YAG:Ce powder produced by CTU in Prague.

c NaOH [mol.dm^{-3}]	Scintillation yield of YAP:Ce [%]				Scintillation yield of YAG:Ce [%]				Scintillation yield of ZnO:Ga [%]			
	^{241}Am	^{239}Pu	^{232}U	^{90}Sr	^{241}Am	^{239}Pu	^{232}U	^{90}Sr	^{241}Am	^{239}Pu	^{232}U	^{90}Sr
0.05	88.7	74.0	95.8	30.2	32.7	37.9	20.5	23.9	43.4	26.3	7.1	4.7
0.1	96.1	79.4	72.3	29.4	30.1 *81.8**	39.7 *67.1**	14.5 *41.6**	22.9 *12.8**	36.6	26.2	4.6	4.2
0.5	96.8	75.0	37.1	27.0	31.9	12.3	5.1	17.5	26.0	3.0	2.8	4.6
1	98.8	74.9	31.2	18.8	38.9	7.9	4.8	17.7	23.5	4.6	2.9	4.9
1.5	92.7	64.5	12.3	18.1	17.8	3.6	5.7	16.8	36.8	5.6	3.6	6.4
3	89.4	40.0	10.2	18.5	24.7	3.8	6.3	14.8	15.9	0.9	2.2	9.5
5	89.7	13.3	11.5	17.3	30.1	1.5	3.7	11.3	0.3	0.1	2.1	10.4
10	82.2	4.4	7.7	9.9	25.3	1.5	3.5	8.5	0.3	0.1	2.9	10.4

* *YAG:Ce$_{CTU}$ was used for comparison*

Sedimentation

Because the time needed to sediment the powder is very long in comparison with the sorption processes, the artificial sedimentation through centrifugation was investigated.

- Etalon - scintillation efficiency of ^{241}Am at 6th - 96 %.
- RPM > 3000 and time > 4 minutes → absolute efficiency (99 ± 1.5) %

Rotation [rpm]	Time [min]	Eff [%]	σ [%]
1000	1	96	2
2000	1	96	1
3000	1	98	2
	2	98	1.5
	4	99	1.5

Influence of types of hydroxides

- NaOH vs. different types of hydroxides.
- Am, Pu and Sr exhibited the same or worse properties when using all investigated powders.
- Only uranium exhibited higher scintillation yield in orders of tens percent (up to 48 % for YAG:Ce and sat. $Cd(OH)_2$) for all powders when using saturated solution of **barium**, **calcium** and **cadmium** hydroxides.

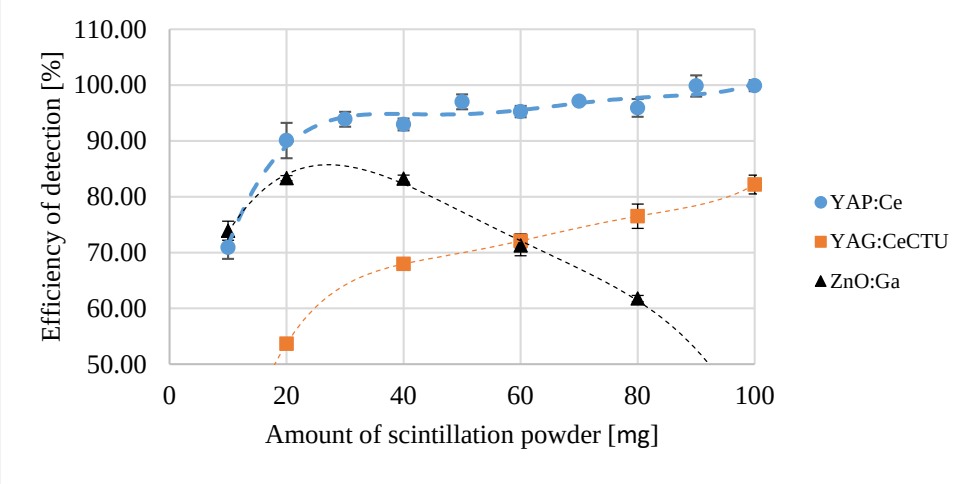
^{232}U									
Etalon ^{232}U + 15 ml 0.1M NaOH	Counts per 40 s			cnts.s^{-1}	σ cnts.s^{-1}	Yield [%]	σ [%]	YDAS*	σ_{YDAS}
	183	168	185	4.5	0.2	15.6	0.8		
0.1M LiOH	181	195	201	4.8	0.2	32.8	1.7	17.2	1.9
0.1M KOH	200	191	205	5.0	0.1	34.1	1.2	18.5	1.5
sat. $Ba(OH)_2.H_2O$	251	179	184	5.1	0.8	35.3	6.8	19.7	6.9
sat. $Cd(OH)_2$	347	323	351	8.5	0.3	63.6	2.6	48.0	2.7
sat. $Ca(OH)_2$	312	315	330	8.0	0.2	59.2	1.6	43.6	1.8
sat. $Bi(OH)_3$	191	136	142	3.9	0.6	25.3	5.1	9.7	5.2
$Bi(OH)_3$ 10 x dill.	225	138	106	3.9	1.3	25.3	10.5	9.7	10.5
$Al(OH)_3$ in 1M NaOH	120	112	105	2.8	0.2	16.1	1.3	0.5	1.5

* Yield deviation against standard

The amount of the scintillator and the volume of solution

- The detection efficiency showed increasing tendency when increasing the amount of YAP:Ce and YAG:Ce powder until the plateau in the range of 90 - 100 mg where the efficiency was (99 ± 1) % or (83 ± 2) % respectively.

- ZnO:Ga shoved different behavior, the maximum plateau was in the range 20 - 40 mg with detection efficiency up to (83 ± 1) %.



Dependence of the efficiency of detection of ^{241}Am on the amount of scintillation powder under 0.1 mol.dm^{-3} NaOH solution.

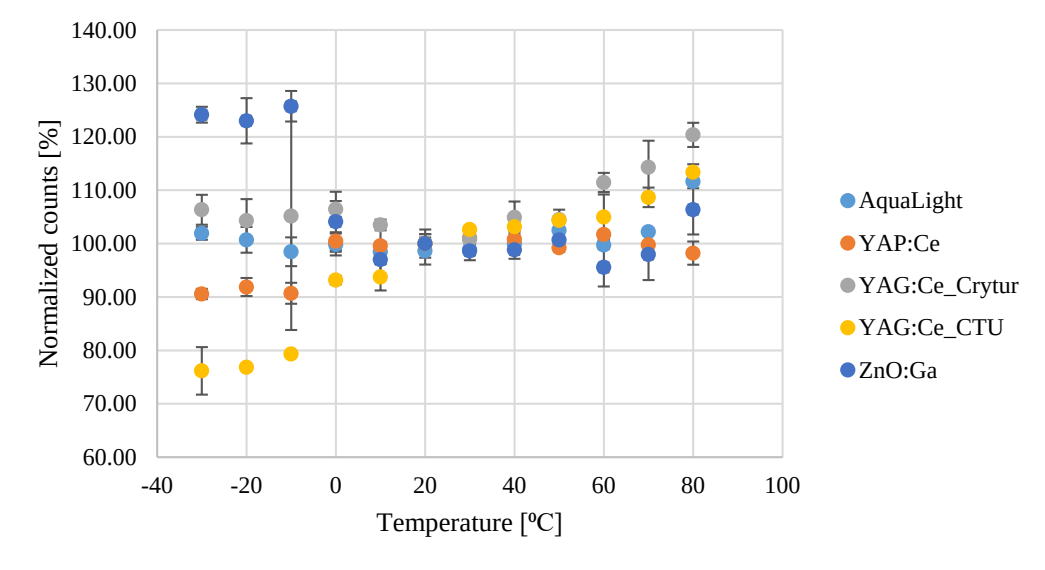
The amount of the scintillator and volume of solution

The volume of solution - no or very little influence at all;

→ the scintillation powders can work from zero volume till full capacity of the scintillation vial without degradation of efficiency, but with slight worsening of uncertainty of the measurement

Volume of solution [ml]	Yield [%]	σ [%]
3	102,2	2,7
6	99,4	1,5
9	97,9	2,6
12	99,2	2,1
15	97,4	2,3

Temperature influence



Temperature dependence of selected scintillation media; observed counts at different temperature were normalized to the value obtained at 20 ºC for each scintillator.

Afterglow

Scintillator	Count rate increase [%] at a wavelength of		Relaxation time [s] for 254/366 nm
	254 nm	366 nm	
Aqualight	10	4	30/20
YAP:Ce	40	9	10/10
ZnO:Ga	23	14	20/20
YAG:Ce Crytur	237	279	-/-*
YAG:Ce CTU	49	49	300/300

** The count rate increase still exhibited 43%, respectively 55%, increment after 600 seconds.*

Redox agents influence

- Reducing agent (1 mmol.dm^{-3} $TiCl_3$).

- YAP:Ce powder:
  - (9 ± 2) % increase for plutonium.
  - Uranium - no significant effect.

- YAG:Ce powder:
  - (39 ± 2) % increase for americium.
  - (12 ± 2) % for plutonium.
  - (62 ± 3) % for uranium.

- ZnO:Ga powder:
  - (38 ± 2) % for americium.
  - (35 ± 2) % for plutonium.
  - (16 ± 1) % for uranium.

Redox agents influence

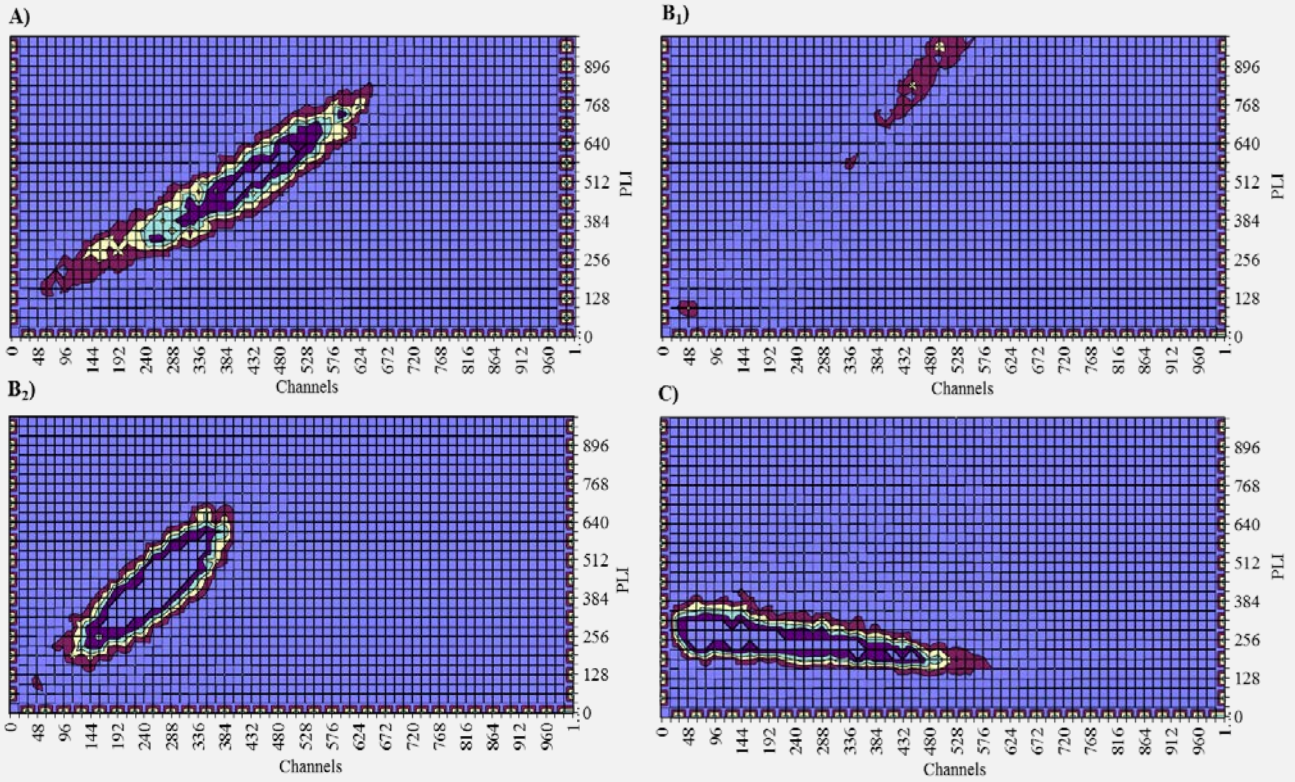
- Oxidizing agent (0.1 and 0.7 mol.dm^{-3} $NaNO_2$)

- YAP:Ce powder:
  - The higher concentration → the worse efficiency was observed.
  - (-12 ± 3) % for Am, (-21 ± 4) % for U, Pu unaffected.

- YAG:Ce powder:
  - slight increase for all nuclides especially at lower concentration.
  - Higher concentration – eff increase only for Am, Pu; U – slight worsening.

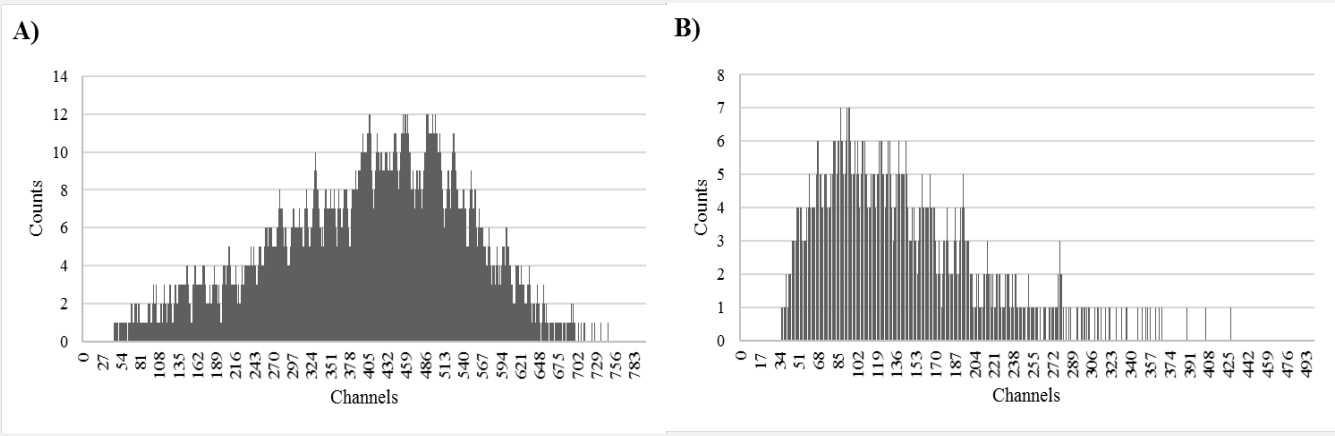
- ZnO:Ga powder
  - The higher the concentration the worse efficiency was observed.
  - Only Am exhibited slight increase (9 ± 2) %.

Shape of the spectra



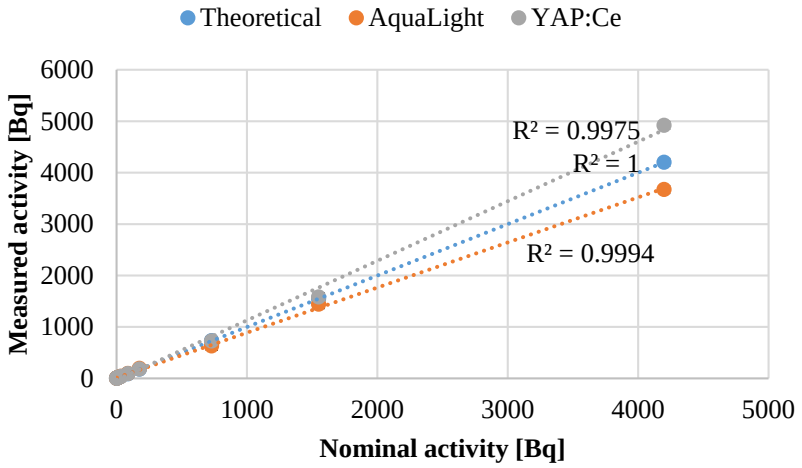
2D spectrum of ^{241}Am using YAP:Ce (A, 0-30 µm), YAG:Ce_{Crytur} (B_1, 30 nm), YAG:Ce_{CTU} (B_2, 30 µm) and ZnO:Ga (C, 5 µm) powder.

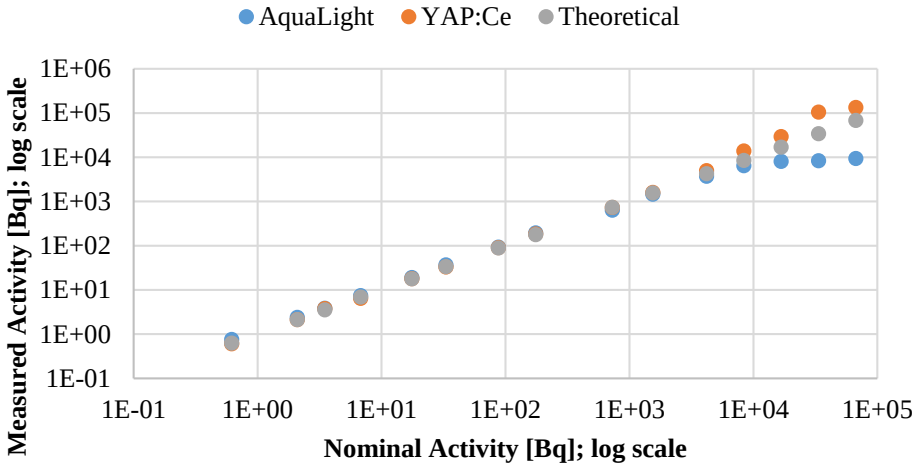
Shape of the spectra – alpha / beta separation



Spectrum of ^{241}Am (A) and ^{90}Sr (B) using YAP:Ce scintillation powder.

Linear dependence of the activity

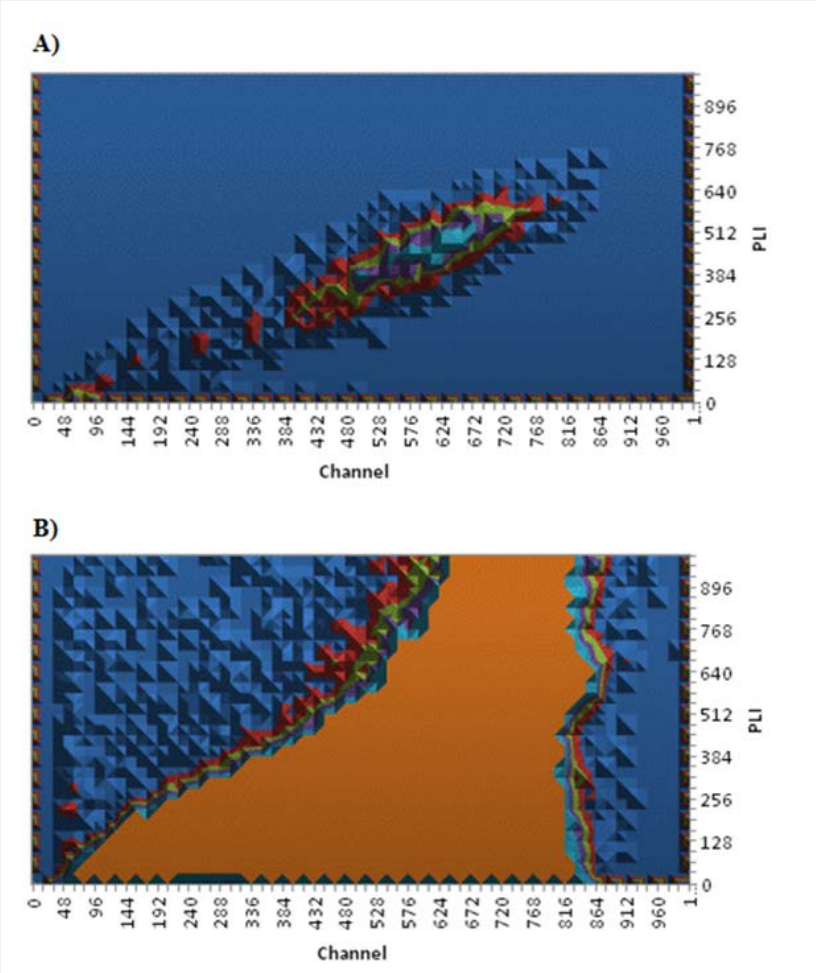


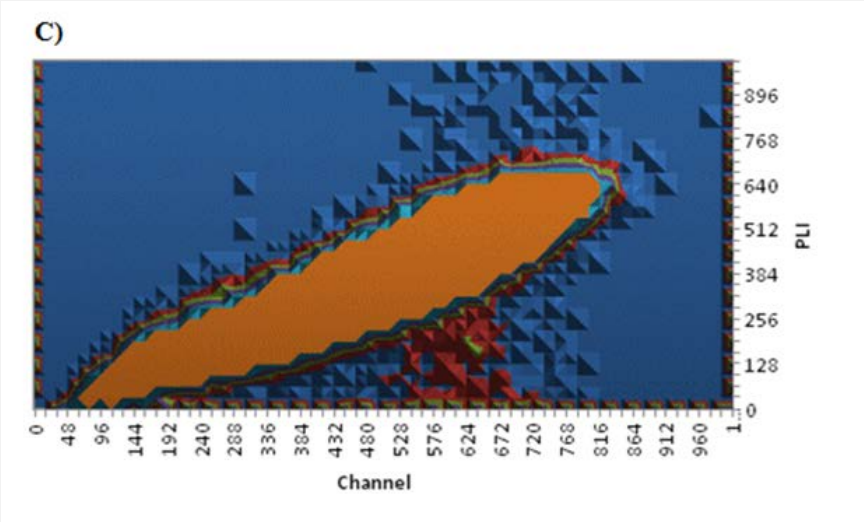


The Comparison of linearity response of AquaLight and YAP:Ce powder to alpha (^{241}Am) particles.

The comparison of nonlinear response of AquaLight and YAP:Ce powder to high activities of samples.

Linear dependence of the activity





The comparison of ^{241}Am spectra with activity of 18 Bq (A) and 67 kBq (B) obtained from Triathler when YAP:Ce powder is used as scintillator media. The (C) shows the count rate increment in range of 500 - 700 channels possible caused by gamma ray interaction ($A_{Am\text{-}241}$=1680 Bq).

Conclusion & further research

- Utilization of solid scintillators (SS) in LS technique is possible.
- Under specific condition SS can fully substitute the liquid cocktail (LC).
- Waste generation is very low in comparison with LC.
- The smaller grain size the better distinction between alpha and beta particles
  → improving the shape of the spectra.

➢ Continue to research suitable scintillators for substitution of liquid cocktail.

➢ Cooperation with Department of Nuclear Chemistry (DNC) of the Czech Technical University in Prague (CTU) on development of new material (Radiation-induced preparation of advanced nanomaterials).

merci
thank you
danke
obrigado
gracias
go raibh maith agat
спасибо
dziękuję
謝謝
chokrane
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terima kasih
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