*Proposal for a project "**R&D of Radiopure Scintillators**"*
Fedor Danevich, INR Kiev, 25 September 2008, after discussions at the RPSCINT 2008 workshop, and further discussion with the workshop participants (H.Kraus, V.Mikhailik, P.de Marcillac, S.Pirro, M.Korjik, A.Dossovitsky, A.Mikhlin, L.Nagornaya, V.Slegel, D.Grigoriev, G.Kovtun, G.Stryganyuk, I.Solsky, D.Spassky, A.S.Nikolaiko, V.I.Tretyak, V.V.Kobychev)

R&D to produce radiopure scintillators for cryogenic dark matter and double beta decay experiments

Duration: 3 years

Cryogenic dark matter and double beta decay experiments require several crystalline scintillators, exhibiting extremely good radiopurity and high light output. There is an excellent opportunity for the production of raw materials, growth of crystals and their testing, R&D on new scintillators and the improvement of existing scintillation materials in the Ukraine, Russia and Belarus. At present the most promising cryogenic scintillators with high light output for dark matter search are $ZnWO_4$, $CaWO_4$, $CaMoO_4$, CaF_2 and BGO. The most promising scintillators for double beta decay experiments are $CdWO_4$ (^{116}Cd, ^{106}Cd), $CaMoO_4$, $PbMoO_4$, $Li_2Zn_2(MoO_4)_3$ (^{100}Mo), ZnSe (^{82}Se). R&D on the scintillation materials Li_2MoO_4, $ZnMoO_4$, $MgWO_4$, that are promising for dark matter and double beta decay search is in progress, as is the further optimization and improvement of $CaWO_4$, $CaMoO_4$, $CdWO_4$, Al_2O_3, LiF, ZnSe, $PbWO_4$, $PbMoO_4$.

The project proposed here could comprise 3 tasks:

1. Development of new and improvement of existing cryogenic scintillators
2. Improvement of radiopurity
3. Testing of radiopure scintillators

1. Scintillators

1.1. Synthesis of promising compounds (poly-crystalline) and their test at low temperature. The most promising compounds should be used for crystal growth and subjected to further investigation and optimization.
1.2. Study of luminescence and scintillation properties down to very low temperatures provides important insight, beneficial towards improving cryogenic scintillators.
1.3. Analytical instrumentation is necessary to control chemical impurities in raw materials with sensitivity at the level of ~0.1 ppm. This is especially important for transition metals.
1.4. Study of deep purification of raw materials on light output of scintillators at low temperatures. It should be emphasized that scintillator performance could benefit from the deep purification required to address the radiopurity issue.

2. Radiopurity

Dark Matter (CRESST, EURECA) and double beta decay experiments call for scintillation materials with extreme radiopurity. Levels of radioactive contamination should not exceed ~10 micro-Bq/kg, which will require significant improvement compared to the present level (0.1 – 100 milli-Bq/kg). $ZnWO_4$ is a good example of a radiopure scintillator (~0.2 milli-Bq/kg level). Still, ~20-fold improvement is needed for EURECA; and that represents a significant challenge. One to three orders of magnitude progress in development of radiopure $CaWO_4$, $CaMoO_4$, BGO, CaF_2, $CdWO_4$ has been demonstrated already, however further improvement by a factor of >100 is necessary. As a first step in the project the direction should be as follows:

1. Deep purification of raw materials is supposed to be the most important issue that needs addressing. Metal purification by vacuum distillation, zone melting, and filtering are very promising approaches, while further study is necessary for the purification of Ca, Li, Se in order to achieve the required low levels.

2. Two to four step re-crystallization, involving inspection and assessment of the produced scintillators after each step.
3. Screening at all stages through ultra-low background γ, α, β spectroscopy is needed in the production of compounds for crystal growing (choice of raw materials, quality control of purified elements and compounds).

All work should be done using highly pure reagents, lab-ware and water. All chemistry should be done in a clean room, and, as far as possible, in nitrogen atmosphere. Careful protection from radon should be foreseen.

3. Testing

1. The low-background scintillation measurements are currently the most appropriate methods of examining the performance of scintillators.
2. Final testing of scintillators involves their operation as low-background cryogenic detectors.
3. R&D of ultra-low-radioactive instrumentation with a sensitivity at the level of 10 micro-Bq/kg (both at room and cryogenic temperature) is necessary.

In short, we propose the following workplan for the project with indication of possible contributors. We estimate cost of the R&D as much as ~2-3 Meuro per 3 years.

Task	**Sub-task**	**Institution / Company**
Scintillators	Synthesis of promising compounds in form of poly-crystals	Kiev University INR, Kiev NeoChem
	Test of poly-crystals at low temperatures	Oxford University INR, Kiev Kiev University Moscow University Lviv Center of Low Temperatures
	Growth of single crystals of promising materials	Crystal producers
	Study of luminescence and scintillation properties of crystals at low temperatures with the aim of obtaining important insight, relevant to improving cryogenic scintillators.	Oxford University INR, Kiev Kiev University Moscow University Lviv Center of Low Temperatures
	Study of deep purification of raw materials, especially its effect on light output of scintillators at low temperatures.	Crystal producers INR, Kiev Oxford University Moscow University Lviv Center of Low Temperatures
	Analytical instrumentation to control chemical impurities in raw materials with a sensitivity of ~0.1 ppm mainly for transition metals.	NeoChem, Laboratori Nazionali del Gran Sasso (LNGS)
	Test of crystals as absorber in scintillating calorimeters	Orsay (Institut d'Astrophysique Spatiale) LNGS

Pure raw materials	Selection and tests of γ, α, β radioactivity of materials of different origin (beginning from ore deposits)	NeoChem – INP Minsk Crystal producers[1] INR, Kiev Modane Canfranc
	Deep purification of initial metals (Zn, Mo, W, Pb) by distillation, zone melting, and recrystallization	KIPT, Kharkov NeoChem IIC Novosibirsk CARAT
	Purification of Ca, Se, Li: organic compounds, recrystallization of CaF, $CaCo_3$, Se and Li	NeoChem IIC Novosonibirsk ISMA Kharkov CARAT
	Control of chemical purity by Röntgen Excitation Analysis (~10-100 ppm), ICP-MS (~0.1-1 ppm), Atom Adsorption Sp. (~1 ppm)	NeoChem European collaborators
	Test for γ, α, β radioactivity of raw materials, compounds, and equipment	INR, Kiev Modane Canfranc
Radiopure crystals	Study on how crystal growth, annealing and machining affect radioactive contamination. Screening of some materials: zircon ceramic, iridium crucibles. Development of radiopure polishing procedures	Crystal producers INP, Minsk INR, Kiev Modane Canfranc
	Crystal production	CARAT ISMA Kharkov IIC Novosibirsk
Test of crystals	Test of optical and scintillation properties	Crystal producers INR Kiev INP Minsk
	Low-background measurements (0.1-1 mBq/kg)	INR Kiev Canfranc Modane
	~10 micro-Bq/kg needs further R&D	INR Kiev, Canfranc, Modane
	Test of luminescence of scintillators at low temperature	Moscow University Kiev University Oxford University Lyon Orsay
	Low-background low-temperature test	Orsay in Canfranc

[1] IIC Novosibirsk, ISMA Kharkov, CARAT