Experience on Solving Issues of Solid Liquid Separation at Cleaning LRW with Considerable Contents of Different Origin Sludge

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ABSTRACT

The report is an overview of totals of the exploratory work having been conducted during several years. A conceptual approach to solving issues on processing so-called "problematic" liquid waste with a considerable quantity of insoluble suspended particles is discussed. Different variants of apparatus for concentration and separation of solid phase (slurry or even sludge) from liquid waste have been developed and laboratory tested. Some results of tests are given in the report.

BACKGROUND

Apart from inorganic salts and radionuclides, most liquid radioactive waste (LRW) can include considerable amounts of insoluble suspended particles, oil emulsions, dissolved surfactants, chelating agents, etc. The majority of the specified admixtures causes severe influences on the sorption cleaning and reduces the LRW decontamination factor. Large insoluble particles and emulsions block the filtering surface of sorbents causing a decrease of sorption capacity and process kinetics as well as increase the hydraulic resistance in a sorption filter because of reduction of a free space between sorbent particles. Dissolved organic substances can essentially decrease the sorption capacity of sorbents due to blocking ion-exchange groups. They can also form organic complexes with radionuclides. These complexes cannot be absorbed and even adsorbed by sorbents. Oxalate-ions, citrate-ions, EDTA and many others are examples of such organic substances.

Current practice of liquid waste processing shows that none of commonly applied methods such as evaporation, settling, filtration, membrane methods can be universal due to their certain disadvantages in all cases. Nevertheless combination of the methods can facilitate the liquid waste processing in the whole.

Membrane methods (ultrafiltration, nanofiltration, reverse osmosis) allow to reach very deep purification of LRW from insoluble admixtures when LRW are pumped through a membrane with an average size of pores less than 1000 Å (or 0,1 μm). Sometimes such result can be achieved by using a microfiltration membrane (sizes of pores 0,1- 10 μm) due to a gel-layer formed. The gel-layer can entrap very fine admixtures in the same way as fine-porous membranes do.

In all cases the gel-layer brings about blocking the free space of membrane pores and increasing the hydraulic resistance of the whole filtering layer as the hydraulic resistance of the gel-layer combines with the one of the membrane. The tangential filtration (or cross-filtration) is usually applied for reducing the thickness of the gel-layer, but even in that case a periodic cleaning (more often a reagent washing-up) of the membrane surface is required. That is why the membrane methods are rarely used for cleaning solutions with high contents of the admixtures (more than 10 g/l).

CONCEPTION

Developing a technology for treatment of liquid waste containing considerable quantities of insoluble substances requires elaboration of a certain concept. Fig.1 is a functional chart of such liquid waste treatment.
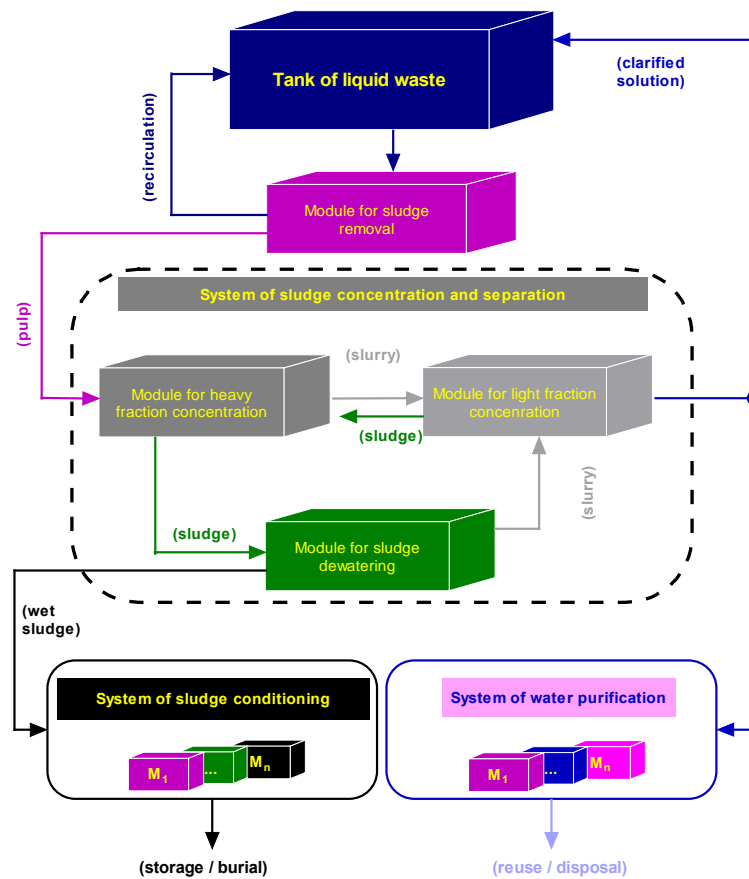

Fig.1. Sludge-containing liquid waste treatment $M_1 - M_n$ – modules or installations

The functional chart should be put in the basis of the treatment of liquid waste containing sludge of any origin. As far as the concept is under development, only the content of the functional chart will be explained hereafter.

The functional chart includes several systems consisting of modules. Each separate module is a completely self-contained installation, which the functionality is based on one or few methods. Such installation includes the following: one or few main apparatus; auxiliary apparatus providing the regular performance of main apparatus; diverse equipment; a common tray; a support frame for fixing the equipment and for loading-unloading and transportation of the module. Each system consists of one or few modules intended for a certain purpose in the system. In the whole, such way of LW treatment allows flexibly approaching to solving the problem of sludge treatment in particular cases.

THEORETICAL

Earlier in the 2005 report [*ICEM'05: The 10th International Conference on Environmental Remediation and Radioactive Waste Management September 4-8, 2005, Scottish Exhibition & Conference Centre, Glasgow, Scotland "OXIDATION-MEMBRANE METHOD FOR CLEANING LRW WITH CONSIDERABLE CONTENTS OF SUSPENDED PARTICLES, OILS AND OTHER POSSIBLE ORGANIC SUBSTANCES" Yury V. Karlin, Vadim A. Ilin, Sergey A. Dmitriev, Alexander V. Gorbachev, Euvgeny S. Volkov*] (Fig. 2–3) there was experimentally shown that introduction of oxidizing agents into aqueous solutions (including LRW), containing considerable quantities of suspended particles, oils and other organic substances, allows to increase the lifespan of micro-or ultrafiltration membranes. Introduction of oxidizing agents into LRW as a rule causes destruction of only organic substances. Oxidation of inorganic substances in aqueous solutions mainly leads to the sludge formation increase.
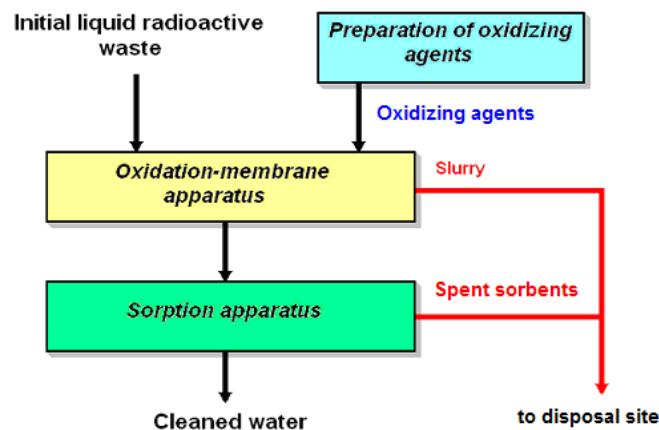

Fig.2. Flow diagram of oxidation-membrane-sorption technology for LRW treatment

Fig. 3. Dewatering oil sludge

To increase the lifespan of membranes being applied for filtration of LRW containing considerable quantities of suspended inorganic particles, the cross-flow filtration can be used. In this process the LRW flux is parallel to the membrane surface. The thickness of residue (or gel-layer) is stabilized at some utmost value because particles formed are permanently slipped away with the LRW flux (Fig. 4).
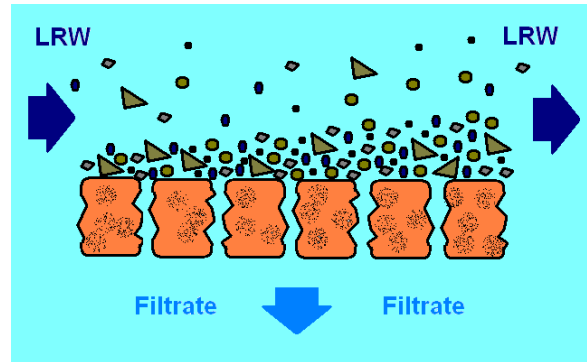

Fig. 4. Cross-flow filtration

The less concentration of particles in the flux and the more velocity of the flux above the membrane surface, the less thickness of the residue layer. The maximum velocity of the flux (more than 10-20 m/s) can be achieved in membrane centrifuge (Fig. 5) [Federal State Unitary Enterprise "Krasnaya Zvezda", Moscow, Russia]. Another way to minimize the thickness of residue is its mechanical removal, e.g. with a scraper or a brush. Some information on such device - disc vacuum filter (Fig. 6) was given in the report [*WM'05 "HLW, TRU, LLW/LW, Mixed, Hazardous Wastes and Environmental Management" February 27 – March 3, 2005 Tucson, Arizona "Development of technology for different nature dredges concentration and separation from LRW." Vadim A. Ilin, Yury V. Karlin, Sergey A. Dmitriev, Euvgeny S. Volkov, Alexander V. Gorbachev*].
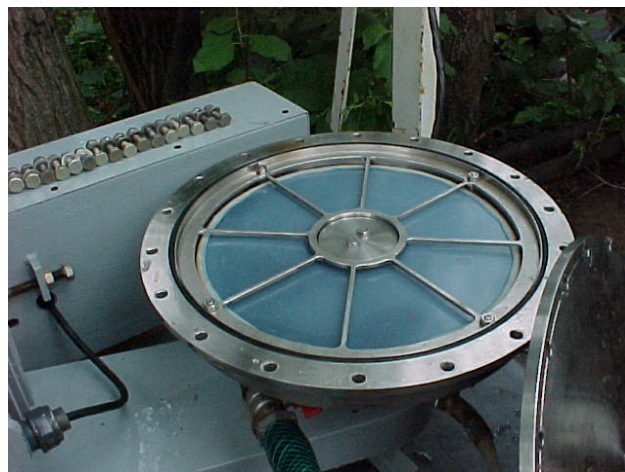

Fig. 5. Overview of "Zolushka" membrane centrifuge (FSUE "Krasnaya Zvezda")

Fig. 6. Overview of "Augean" disc vacuum filter (S $_{filtering\ area}$=2 m^2)

Common disadvantage of the membrane centrifuge and the disc vacuum filter is an abrasive impact of residue on the filtering surface because of its permanent movement (slipping away, discharging or being cut with scrapers) on the filtering surface. At the same time, the presence of hard abrasive particles in LRW (metal, concrete and etc.) is a usual case. Besides, incomplete removal of the residue from the filtering surface in time causes its compaction and it blocks the membrane filtering surface. Further, there will be discussed a filtration or membrane apparatus in which the abrasive impact of residue is minimum.

The apparatus mode of operation (Fig. 7) is traditional. Aqueous solution or LRW spins around a pintle. Centrifugal force drops suspended particles (with density higher than the one of water) onto the walls of the apparatus, where they scrawl as residue down to the conic bottom. The apparatus is similar to a hydrocyclone and a microfiltration filter combined together. The main feature is that filtrate outcomes from a rotated cartridge-type filter (microfiltration or ultrafiltration). In the hydrocyclone the treated solution is discharged through a socket located on the pivot pin (without filtration). As distinct from the hydrocyclone, in the cartridge-type microfiltration apparatus all the particles are transported to surface of the filter because of absence of the liquid rotation (Fig. 7a). It should be noted that the filtration condition essentially change in such apparatus. Due to change of the rotation rate, it is possible to change the residue structure on the filtering surface (Fig. 7b-7c).
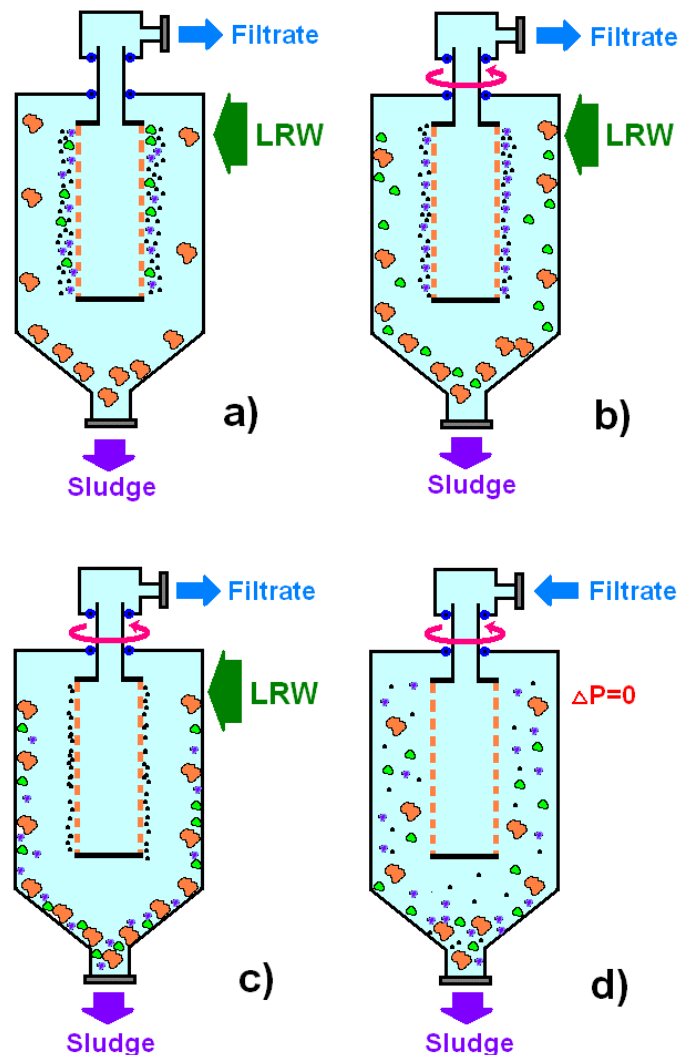
Fig. 7. Mode of operating the filtering apparatus with the rotary filter:
a)absence of rotation – ordinary cartridge-type microfilter; b) low rate rotation (only coarse particles are thrown away); c) high rate rotation (the majority of particles are thrown away); d) absence of overpressure in the apparatus – reverse flushing of the filtering surface occurs.

Regeneration of the filtering surface in the filtration apparatus with the pintle rotary membrane filter becomes easier. To do this, it is necessary just to switch off overpressure inside the apparatus and to cease LRW feeding. The filtrate under action of the centrifugal force is coming back into the apparatus through the filtering membrane and flushing the residue from the surface of the rotary membrane filter into the apparatus inner volume (Fig. 7d).

To estimate the minimum size of a particle thrown away from the membrane filtering surface into the apparatus inner volume, the following main assumptions should be taken:
- Particle resistance force to the incoming flux can be evaluated by the Stokes Law (laminar flow regime);
- Particle is sphere-shaped;
- Mutual influence of particles is not taken into consideration;

- Relative motion of particles in pintle and tangential directions is not considered;
- Angular velocity of the particle rotation around the filtrate pin is equal to the one of the filtrate.
-

Let's use the equality of the resistance force to motion of the particle being on the outer surface of the filter to the resultant force and pressure force

$$F_C - F_W = F_{St} \tag{Eq. 1}$$

$$(\rho_P - \rho_W)\frac{\pi d_P^3}{6}\omega^2 R = 3\pi d_P \mu w \tag{Eq. 2}$$

here ρ_P, ρ_W – density values for the particle and water; d_P – particle diameter; ω – angular velocity of filter rotation (radian/s); R – radius of the outer surface of the filter; μ – water viscosity; w – water flux velocity relative to the particle in the radial direction, which can be find by equation:

$$w = \frac{Q}{2\pi R H} \tag{Eq. 3}$$

where Q –the apparatus volume productivity by filtrate, H – height of the filter.

From equations (2) and (3), it is easy to obtain the equation for the minimum size of the particles, which will not participate in formation of the residue (gel-layer) on the cylindrical surface of the filter having the radius R:

$$d_P = \frac{1}{\omega R}\sqrt{\frac{9\mu Q}{\pi H(\rho_P - \rho_W)}} \tag{Eq. 4}$$

or (for the angular velocity of the filter rotation in rpm, n=30ω/πR, and specific productivity of the filtration apparatus, q (m/s)=Q/2πRH):

$$d_P \approx \frac{40,5}{n}\sqrt{\frac{\mu q}{R^3(\rho_P - \rho_W)}} \tag{Eq. 5}$$

Let's find d_P at initial figures: $\mu = 10^{-3}$ Pa·s, $\rho_P = 1,5 \cdot 10^3$ kg/m³, $\rho_W = 1,0 \cdot 10^3$ kg/m³, $R = 0,03 \div 0,2$ m, $q = 1,4 \cdot 10^{-4}$ m/s (or 500 l/m²·h), $n = 0 \div 5000$ rpm. Results of calculation (Fig. 8) show that the radius of the filter and its rotation velocity considerably influence the size of particles forming the residue (gel-layer) on the filtering surface.
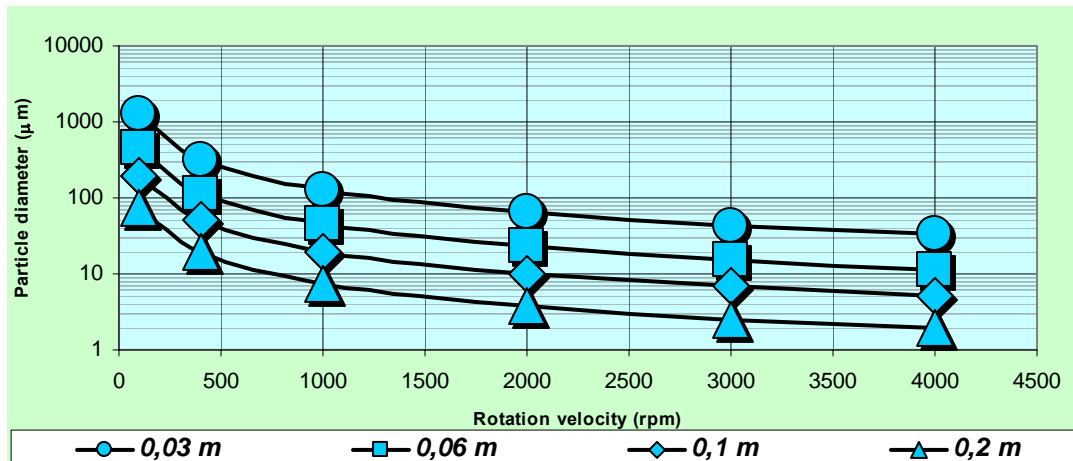
Fig. 8. Dependence of minimum particle size on rotation velocity of the filter

EXPERIMENTAL

A draft of apparatus with rotary cartridge-type microfilter (conventional name - "VIKING") is given in Fig. 9.
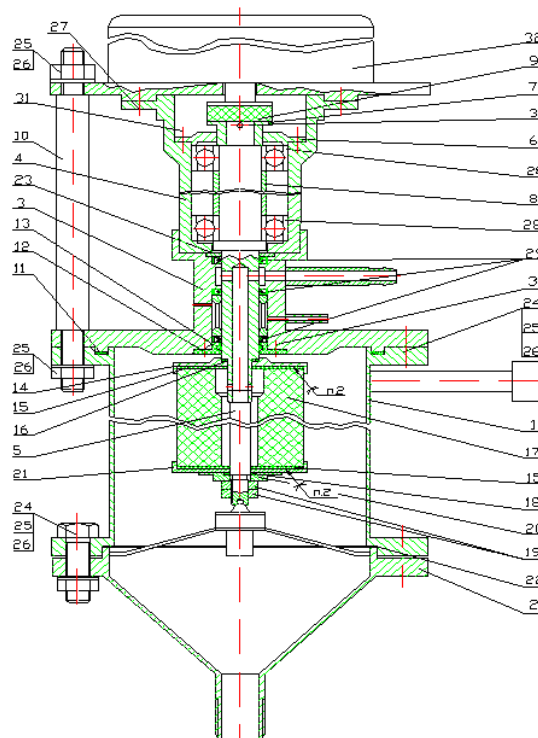

Fig. 9. The "VIKING" filtration apparatus with rotary cartridge-type microfilter:
1 - case, 2 - bottom, 3 - lid, 4 – case of bearing unit, 5 - drive shaft, 6 - cover, 7 - clutch, 8 - ring, 9 - crossbar, 10 - stud, 11 - gasket, 12 - bush, 13 - sealing gland, 14 - flange, 15 - gasket, 16 – gasket, 17 – cartridge-type microfiltration polymeric element, 18 - gasket, 19 – nut, 20 - washer, 21 – flange, 22 – gasket, 23 – washer, 24÷27, 31 – fixing wares, 28 - bearing, 29 – sleeve gasket, 30 - dowel, 32 - electric motor

The overview and hydraulic circuit of the "VIKING" pilot plant are in Fig. 10 – 11 accordingly.

Fig. 10. Overview of "VIKING" apparatus and installation
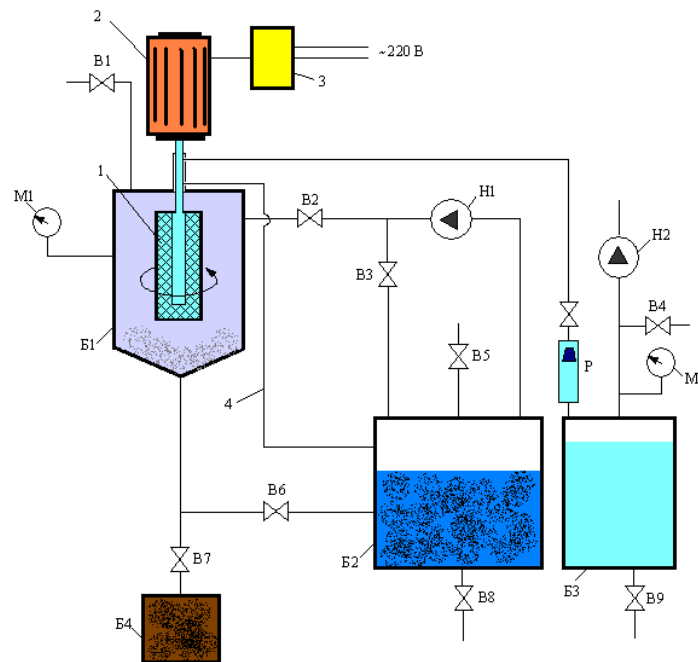

Fig. 11. Hydraulic circuit of "VIKING" installation:
1 – cartridge-type filtration element; 2 – electric motor; 3 – control unit; 4 – leakage discharge pipeline for apparatus Б1; Б1 – case of filtration apparatus; Б2 – tank for treated LRW, Б3 –tank-receiver for filtrate (can be under vacuum), Б4 –tank-collector for sludge; В1…В9 – valves; Р - rotameter, М1 – pressure-gauge; М2 – vacuum-gauge; Н1 – rotary pump; Н2 – vacuum pump

The installation operating mode is follows. Pump Н1 circulates the treated solution between tank Б2 and apparatus Б1. Using valves В2 and В3, the surplus pressure of 0,02-0,04 MPa is

produced in apparatus Б1. Vacuum pump H2, creating vacuum in tank Б3, is concurrently switched on. The pressure difference produced by pumps H1 and H2 causes filtration of solution (LRW) through the rotary cartridge-type microfilter 1 (in the given case, a polypropylene cartridge-type microfilter EFP-PP-5-500 were used; produced by SIE "Eco-Express", Obninsk, RF, outer diameter of filter - 0,064 m, height - 0,25 m, average size of pores - 5 μm). Sludge is collected in the conical bottom and periodically unloaded into tank Б4 or backward into tank Б2 (concentrating regime).

Trials of the installation on distilled water showed that (Fig. 12) the rotation acceleration causes decrease of the installation because overcoming the centrifugal force requires some additional energy.
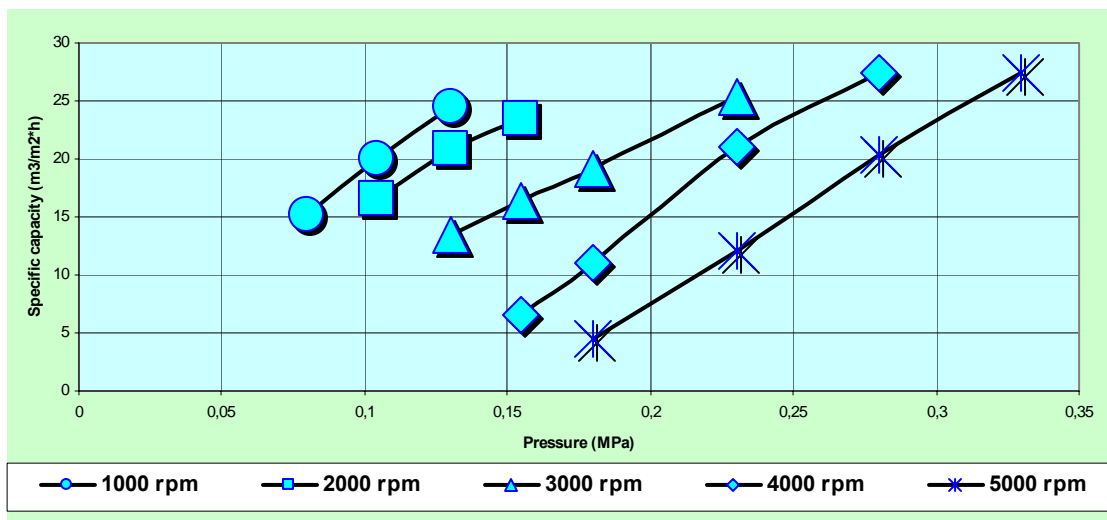

Fig. 12. Installation "VIKING": specific productivity dependence on pressure at different rotation velocity of the cartridge-type microfilter

Further trials were conducted with simulated solutions, containing real silt from operating settler-ponds of the RADON disposal site drainage system. Fractional composition of silt and contamination of fractions with Cs-137 are represented in Fig. 13. It should be noted that unlike sandy grounds, the silt contamination has volumetric nature. It means the sharp increase of specific activity at decrease of the average size of fractions. Accumulation of Cs-137 in silt can be explained by the fact that soils at RADON disposal site are mainly clays, in particular, montmorillonite, which is an excellent sorbent for Cs-137.
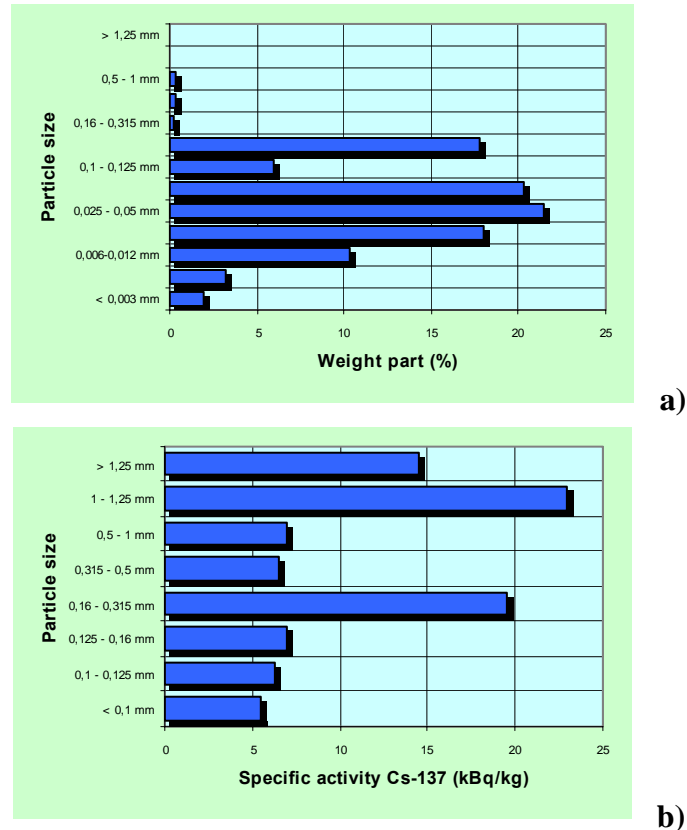
a)

b)

Fig. 13. Dependence of a) weight share; b) specific activity of Cs-137 on silt particle sizes

The first series of experiments was conducted with a simulated silt solution at silt contents about 160 mg/l and particle size – less than 10 μm (sedimentary prepared – the upper discharged flux of the simulated solution in a vessel after 40-50 minutes was taken out). Concentration of particles in the initial solution and in the filtrate was analyzed by gravimetric method (for that, ultrafiltration in a static cell (Fig. 14) was used). Dependence of the particle carry-over through the cartridge-type microfilter on its rotation velocity at constant value of filtrate production (about 150 l/h) was investigated.
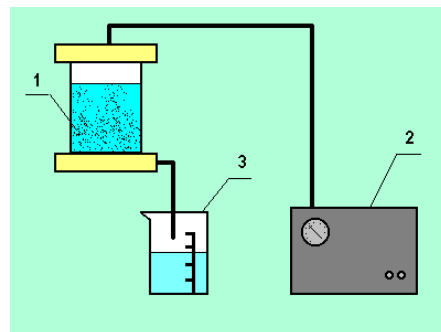

Fig. 14. Ultrafiltration static cell stand:
1 – ultrafiltration cell, 2 - compressor, 3 - graduated cylinder

Results of the experiments (see Fig. 15) show that acceleration of the filter rotation causes decrease of the particle contents in the filtrate. Thus, the influence of centrifugal force on more dense particles in aqueous solution is higher than on water. The efficient coefficient of particle diffusion in the labyrinth of porous space of the microfilter decreases.
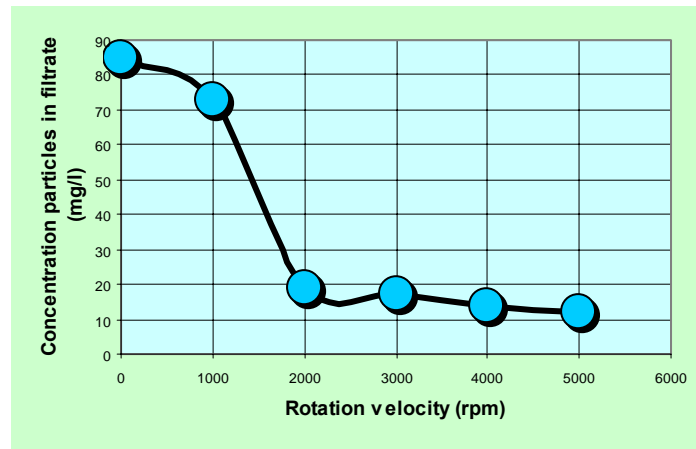

Fig. 15. Dependence of particle contents in filtrate on rotation velocity of the cartridge-type microfilter (productivity 150 l/h)

The next experiments were carried out with the same simulated solution of silt (160 mg/l, particle size less than 10 µm). Rotation velocity (5000 rpm) of the cartridge-type microfilter was maintained unchangeable, while productivity of the installation by filtrate was changed. Increase of particle contents in the filtrate at increasing the microfilter productivity (Fig. 16) can be explained by the concentration polarization of the microfilter surface on the side of the initial solution. Concentration of particles in the filtered solution near the microfilter surface increases at increase of the flux velocity into the microfilter, hence, concentration of particles incoming into the filter increases.
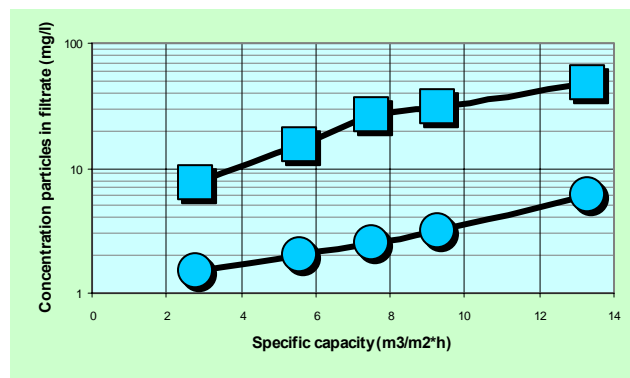

Fig. 16. Dependence of particle contents in the filtrate on productivity of the cartridge-type microfilter (rotation velocity - 5000 rpm) at filtering simulated solution: a) ○,160 mg/l, particle size – less than 10 µm; b) □, 380 mg/l, polydisperse composition

The second series of experiments was conducted with a simulated solution containing contaminated silt (380 mg/l) and the one of a polydisperse composition. The procedure was the same as in the previous series. Results obtained were similar (Fig. 16). Drastic increase of

particle contents in the filtrate at using the polydisperse solution is possibly caused by pounding the silt particles in the centrifugal pump while circulating.

With the microfilter productivity to be decreased by 20%, an attempt was undertaken to regenerate it with peroxide aqueous (about 10%) solution. The washing led to decrease of the filter productivity by another 5%. The filter productivity was restored after its washing with 1 g/l aqueous solution of citric acid during 4 hours at 30 ^{o}C. The result obtained can be explained by the fact that the microfilter had accumulated ferrous products being in plenty in silt.

Lifespan trials of the "VIKING" installation were carried out with a simulated solution of silt at its contents of 90 mg/l and 380 mg/l. Time of hydraulic resistance increase from 0,14 MPa up to 0,26 MPa on the microfilter at the installation productivity by filtrate of 75 l/h and rotation velocity 4000 rpm of the cartridge-type microfilter was determined. The time of pressure increase for the simulated 90 mg/l silt solution was 60 min, and for 380 mg/l silt solution – 35 min. The same trials with the 90 mg/l silt solution were done in the static regime (no rotation of the cartridge-type microfilter). The time of pressure increase made only 3 min.

Currently, the possibility to replace the polymeric filter of cylindrical filtering surface (length - 0,25 m, square - 0,05 m^2) for the one of goffer net surface (made of stainless steel, cell size - 10 μm, total filtering square - 0,22 m^2, Fig. 17).

Fig. 17. Overview of cartridge-type net filter

Unlike the polymeric microfilter, the cartridge-type net filter is a surface-action one. Therefore, it will begin properly to operate when a gel-layer forms on its surface. An advantage of the cartridge-type net filter in comparison to the polymeric one is possibility to regenerate it by the reverse washing. Using the simulated solution (silt - 250 mg/l, oil - 45 mg/l, surfactants - 35 mg/l), in static conditions, it was demonstrated that the cartridge-type net filter cleaned water more than 99% from suspension and emulsion (Fig. 18). At that the cartridge-type net filter can be easily regenerated by the reverse washing (Fig. 19).

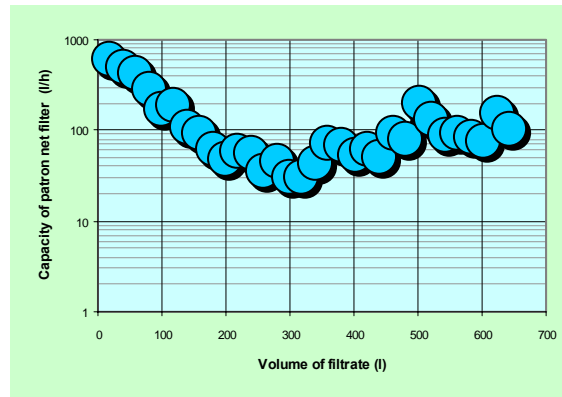Fig. 18. Quality of initial solution cleaning in the cartridge-type net filter

Fig. 19. Dependence of cartridge-type net filter productivity on volume of simulated solution cleaned (regeneration after 502 l, 603 l).

CONCLUSIONS

Investigation and study of possible ways to separate liquid and solid phase are still continued.

Within 2006-2007 it is supposed to do the following work:

1. To finish development of the "VIKING" apparatus applying an optimal filtering element design and material.

2. Experimentally to determine lifespan of the filtering element and washing regimes for liquid waste containing various insoluble contaminants.

3. To develop a generalized technological scheme for treating the LRW with considerable contents of suspension, oils and other possible organic substances.