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ASSESSMENT OF LOESS COMPOSITION AND STRUCTURE IN CONNECTION WITH RADIOACTIVE WASTE DISPOSAL

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ABSTRACT
Investigations have been carried out on the composition and structure of loess in the vicinity of the Kozloduy Nuclear Power Plant in connection with the long-term disposal of radioactive waste. The thickness of loess in the investigated region is in the range from 10 to 40 m. The loess grain-size composition is characterised by predominating silty fraction reaching up to 80%. The clayey fraction content varies from 10 to 20%. The silty and sandy fractions contain mainly quartz, feldspar, mica and carbonates and the clayey fraction – mainly chlorite and smectite. The most important components of the chemical composition of loess are SiO_2, Al_2O_3, Fe_2O_3 and FeO, CaO and MgO. The amount of the water-soluble salts, presented mainly by carbonates (bicarbonates), sulphates and chlorides, is low. The sorption capacity is about 6 meq/100 g. According to the classification of Minkov the loess structure is granular or aggregate-granular, its basic property being the incomplete consolidation and collapsibility. The dry density is about or less than 1.4 g/cm^3, and the degree of water saturation is S_r = 0.30-0.55. The volume of macropores under loading of 0.3 MPa is 6.5 – 7.0%. The filtration coefficient varies from 2.10 $^{-5}$ to 5.10 $^{-6}$ m/s. The properties of loess are considered in the present report from the viewpoint of safety of both existing and future repositories. The main conclusion of the investigations is that regardless of some suitable qualities, loess in its natural state cannot be an efficient barrier against radionuclide migration. It is necessary to apply methods for its compacting and strengthening. In Bulgaria there is experience in the successful application of such methods, as proved by the operation of the Kozloduy NPP.

INTRODUCTION

The beginning of nuclear power generation in Bulgaria dates back to 1974 with the launching of the first reactor of Kozloduy Nuclear Power Plant, as other 5 reactors have sequentially been put into operation till 1991. In compliance with the international practice, the low and intermediate level radioactive waste (LILRAW) released during the plant operation should be subjected to final disposal in surface repositories after their temporary storage. As a result of the whole period of operation of the Kozloduy NPP, the closing of its first two reactors and the eventual operation of the Belene NPP, it is expected that the LILRAW volume in the conditioned state will amount to about 100 000 m^3. These wastes are stored at present in temporary storages on the plant territory and it is envisaged to construct a permanent repository. In this context it is very important that a suitable geological environment for its construction should be selected. The investigations carried out so far both in the country and abroad (Hungary and Romania), prove that loess could be considered as a prospective medium for storing low and intermediate level radioactive waste. This geological medium together with the engineering structure (the repository) plays the role of a barrier against the radionuclide migration and is a basic factor for the long-term stability of the future repository system. The assessment performed further on refers also to the existing repositories on loess at the plant site.

Loess was deposited in an aeolian way during the Quaternary and covered in a mantle manner the existing Pliocene relief. The formed loess horizons alternate with fossil soils (paleosoils), thus forming a common "loess complex". The number of horizons in the studied region reaches up to six, separated by five fossil soils. The following geomorphological forms are outlined: loess plateaux, where loess reaches the thickness of up to 30 –40 m; loess slopes; wadies "inheriting" old dried riverbeds, where the loess cover becomes thinner, as well as loess plates situated in the plateaux. The complex is characterised by the following specific features: changes in the facial composition in both horizontal and vertical direction – from sandy to clayey loess from the north to the south. A similar transition is also observed from the plateaux to the slopes. The loess structure and composition on the wady slopes are altered, the loess being generally more dense and clayey in comparison with the loess of the terraces. Another specific feature is the increased content of clay in the more deeply situated horizons. The upper ones in the region of Kozloduy are classified as sandy or silty (typical) loess while the bottom horizons fall within the group of clayey loess.

Loess composition and structure influence to a great extent its engineering geological properties and especially its behaviour as a soil base. The study of loess composition and structure has several aspects with respect to radioactive waste disposal, namely:

- stability of loess minerals and structure under the influence of climatic factors and groundwater in the course of a long time period;
- effect exerted by the properties and structure on the long-term stability of the soil base of a LILRAW repository;
- presence and effect of chemical compounds in loess, which are aggressive towards concrete, steel and other building materials;
- possibilities for improving loess structure by means of methods for strengthening and compacting.

On the basis of the above mentioned aspects an assessment will be made for the composition and structure of loess in the

Kozloduy NPP proximity form the viewpoint of an eventual medium for LILRAW long-term disposal.

COMPOSITION AND PROPERTIES OF LOESS

Grain-size distribution and plasticity limits

The silty fraction is the basic one in loess. It reaches up to 80% in sandy, silty (typical) and clayey loess, up to 50% - in loess-like sands and up to 60% - in loess-like clays. A clearly expressed trend is observed for the increase of the clayey fraction in both the redeposited loess and in the fossil soils as a result of long-term weathering processes.

The changes in the quantity of the clayey fraction (< 0.005 mm) are used as a criterion in loess classification (Minkov, 1968): loess-like sand and sandy loess, containing up to 10% of clayey fraction; silty loess – up to 20%; clayey loess – up to 30% and loess-like clay - >30%.

The considered region is characterised by sandy and mainly silty loess, to a smaller extent – by clayey loess and to a much smaller degree – by loess-like sand, which is observed along the bank of the Danube River. Redeposited loess has been established too. For this reason, samples of sandy and typical loess from the region of the Kozloduy NPP have been investigated. The tested sandy loess samples (according to the Bulgarian State Standard BDS 2762-75) showed the following composition: 10% of sandy fraction, 84% of silty fraction and 6% of clayey fraction. The fraction larger than 2 mm (Table 1) is absent in sandy loess. One variety of silty loess has been investigated, yielding the following composition – 11% of sandy fraction, 68% of silty fraction and 17% of clayey fraction. The amount of the fraction >2 mm is 4%. The silty fraction is the dominating one in both facial varieties but its share is relatively lower in the tipical loess at the expense of the clayey fraction. The uniformity coefficient $u=d_{60}/d_{10}$ is 7.06 and 15.00 respectively. The typical loess possesses higher uniformity coefficient, i.e. it is characterised by complete granulometric curve, which is favourable with respect to loess compaction. The two samples are defined as silty clayey sand according to BDS 676-85.

Mineral composition

Loess contains a great number of minerals, irregularly distributed in the different fractions. The available data from the studies carried out so far (Minkov, 1968; Petrussenko, 1973; Antonov, 2002) are analysed further on.

The predominating minerals in loess are quartz, feldspar, mica and carbonates. The DSC curves of the loess samples from the region are complicated because the reflections of many minerals overlap with each other. Regardless of this fact, strong reflexes are observed for: quartz at 0.333 nm, 0.424 nm, 0.1812 nm and 0.1815 nm, 0.1537, 0.137 nm; calcite – at 0.302 nm; dolomite – at 0.288 nm (dominating quantitatively the calcite in the silty loess); micas – at 0.989 nm and 0.983 nm, 0.496-0.498 nm; feldspars – at 0.319-0.322 nm and 0.318-0.323, with plagioclase being mainly the dominating species and with traces of potassium feldspar, the latter being in a greater amount in the sandy loess sample; clayey minerals – at 0.53 nm, 0.705 nm (chlorite), 0.352 nm and 0.701 nm; smectite (montmorillonite) – at 1.403 nm.
The granulometric fractions of loess are characterised by the following composition:

Fraction >2 mm. It is represented mainly by quartz, single muscovite and biotite scales and single feldspar grains (the light part of the fraction). Bigger carbonate and clayey-carbonate concretion, known as loess dolls, are encountered too. Single grains of amphibole, epidote and ore minerals are seldom observed as representatives of the heavy part of the fraction.

Fraction 2 – 0.01 mm. The greater part of the loess mass falls within this fraction. The light part of the fraction contains mainly quartz, micas – muscovite and biotite, feldspars and carbonates.

Quartz is the basic mineral (more than 50-60%) and is represented by rounded transparent grains of irregular shape.

The micas – muscovite and biotite, occupy the second place after quartz. The muscovite amount exceeds the biotite one from 4 to 8 times and is usually represented by thin colourless platelets. Its content in the total loess mass varies from 15 to 30%. The biotite is observed in the form of small scales and not so often – of oval shaped platelets.

Feldspars occupy the third place but sometimes their quantity might exceed that of micas.

Table 1. Classification indices of the used samples

Loess variety	Particle-size distribution [%]				Solid density	Plasticity			Uniformity coefficient
	> 2 mm	2 - 0,1 mm	0,1 - 0,005 mm	< 0,005 mm	ρ_s g/cm^3	w_L %	w_P %	I_P %	u [-]
Sandy loess	0	10	84	6	2,74	28,0	25,3	2,7	7,06
Typical loess	4	11	68	17	2,72	25,5	20,0	5,5	15,00

They are encountered either as weathered or quite fresh particles of angular, slightly rounded or oval shape. In principle, the feldspars in fossil soils are strongly weathered and their quantity is two to three times lower than in the loess horizons. Orthoclase and seldom microcline are found as representatives of potassium feldspar and albite and oligoclase – of plagioclase.

All these minerals are resistant to the action of the atmospheric factors and eventual infiltrates that might be released by the conditioned RAW.

The carbonate content of loess varies within a broad range both in horizontal and vertical direction – from 3-4% in leached loess to 30% in the carbonate horizons of the paleosoils. This variation can be used as a natural analogue considering migration properties of some radionuclides. A trend of changing the carbonate content in horizontal direction is observed: in loess-like sands – average of 18%, in sandy loess – about 15%, in silty loess – to 17% and in clayey loess – to 13-14%. The present investigations have shown a carbonate content of 22.9% for sandy loess and 16.32% for silty loess (Table 2). The carbonates are represented by calcite and by a very small amount (1-2%) of dolomite. Carbonates are minerals subjected to destruction under the continuous action of atmospheric water, which is displayed by the presence of thick carbonate zones in the buried soils. Their dissolution is increased in acidic medium. The light fraction comprises also the secondarily formed highly water-resistant clayey-carbonate, iron- or magnesium-carbonate aggregates, ferromanganese salts, etc., which are irregularly distributed and in negligible quantities.

The amount of the heavy fraction is smaller and is represented by about 20 minerals, the quantity of the non-transparent minerals being the highest.

Fraction 0.01 - 0.005 mm. According to the microscopic investigations the particles with sizes from 0.01 to 0.005 mm are represented by quartz, altered feldspars and micas, calcite, rarely by single small crystals of heavy minerals and clayey aggregates.

Fraction <0.005 mm (clay). This fraction consists mainly of kaolinite, hydromicas and montmorillonite. Dispersed quartz and dispersed carbonates are also observed. The following trends have been established for the changes of the clayey minerals in horizontal and vertical direction: the finely dispersed fraction of the contemporary soil (loess chernosem) consists of minerals with three-layered crystal lattices, mainly montmorillonite; the clayey fraction of loess contains predominantly minerals of the group of hydromicas (mainly illite) and of montmorillonite; the hydromicas are predominant in sandy loess; montmorillonite is predominant, hydromicas decrease and kaolinite is almost negligible in the direction towards loess-like clay and fossil soils. The samples investigated by us showed the presence of chlorite, kaolinite and montmorillonite (traces).

Humus content – loess always contains a certain amount of humus (0.1 – 0.6%), which is usually included in the clayey fraction. The humus content of the investigated samples is 0.22 and 0.67% (Table 2). The highest humus quantity is observed in the fossil soils. Two types of humus are distinguished – primary and secondary. The primary humus is encountered as a fine incrustation on the phytogenic macropores. The post-sedimentation humus generation is contained in the soils in the form of diverse aggregates.

Chemical composition

The chemical compounds contained in loess can be distinguished in three main groups: of major importance (primary) – SiO_2, Al_2O_3, Fe_2O_3, FeO; of secondary importance – CaO and MgO, and less important – all the rest compounds.

The chemical composition of loess is in accordance with its mineral composition. The quantity of SiO_2 dominates all the other components in all loess varieties and horizons. The silica amount in loess changes within relatively narrow limits – from 58 to 51%, decreasing gradually from the sandy to the clayey facies. The amount of Al_2O_3 + Fe_2O_3 changes in the opposite direction from that of SiO_2, moreover within a significant range – from 13.0% in sandy loess to 19.0% in loess-like clay. The quantities in the buried soils are higher.

The amounts of the earth alkaline oxides (CaO and MgO) change in the reverse direction with increasing the distance from the Danube River – CaO is increased while MgO is decreased. This is in good agreement with the reduction of mica and magnesium carbonate amounts in clayey loess. The sum CaO + MgO in the buried soils is lower due to the leaching of carbonates.

At the same time the soils have lower alkaline oxide (Na_2O + K_2O) amounts and a higher TiO_2 content.

The following data (Minkov, 1968) might be used as an illustration of the chemical composition of the clayey fraction.

Table 2. Chemical and physico-chemical data of the used samples

Loess variety	Aqueous extract 1:10 - 24 h meq/100g							Carbonate content	Humus content	Sorption capacity
	HCO_3^-	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	dry residium [%]	pH	[%]	[%]	[meq/100g]
Sandy loess	0,976	1,0	0,4	0,27	0,17	0,12	8,1	22,4	0,22	5,88
Typical loess	1,31	1,0	1,0	0,11	0,15	0,12	8,0	16,32	0,67	6,20

Three loess samples, collected sequentially from a depth of 0.60, 2.10 and 10.0 m, exhibit the following chemical composition of this fraction (% of the silicate mass without the ignition losses): SiO_2 – 46.44 to 51.08; Al_2O_3 – 20.99 to 15.49; Fe_2O_3 – 12.44 to 19.77; P_2O_5 – 0.48 to 0.24; TiO_2 – 0.74 to 1.18; CaO – 2.44 to 1.52; MgO – 3.42 to 3.35. The SiO_2 content is about 20% lower and the sum R_2O_3 is about 18-20% higher than that in the total loess mass. The quantity of Al_2O_3 is increased significantly in comparison with Fe_2O_3, and the MgO amount exceeds the CaO one.

Water soluble compounds. The total content of the easily soluble and average and hardly dissolved salts determines the type and degree of loess salinity. The loess in the investigated region contains, although in rather small amounts, the three water soluble compounds – carbonates (bicarbonates), sulphates and chlorides (Table 2).

The carbonate (bicarbonate) quantities are the highest. In about 80% of the samples HCO_3^- amounts to more than 60% of the total anion sum. This determines the exclusively carbonate type of salinity. Only the loess-like sands and some sandy loess samples exhibit slightly expressed carbonate-sulphate salinity. The HCO_3^- amount in the first, second and third loess horizon slightly increases from sandy to clayey loess in parallel with the increasing finely dispersed carbonates. The carbonate content is the highest in the Lom district both in loess (up to 4.08 meq/100 g) and in buried soils (up to 9.54 meq/100 g). The carbonate amounts gradually decrease to the east. The HCO_3^- quantity near Kozloduy is 0.976 meq/100 g for sandy loess and 1.31 meq/100 g for typical loess. The loess in the sections of our sampling exhibits also a carbonate type of salinity. The bicarbonate quantity is more than 69% of the total anion sum for the sandy loess sample and more than 83% for the typical loess sample. In general, the chemical composition of the samples is within the framework of the expected values for the region of the Kozloduy NPP.

Sulphates occupy the second place. Their quantity is higher only in sandy loess. Their amounts decrease in eastern and southern the direction and do not exceed 30% of the anion sum for about 80% of the samples. In contrast to carbonates, they decrease in the fossil soils and exhibits maximums at a certain depth below them. The sulphate content in the samples investigated by us is lower, respectively 0.17 meq/100 g for sandy and 0.15 meq/100 g for typical loess (Table 2), which could be considered as a favourable circumstance with respect to the long-term stability of the RAW barriers, using Portland cement.

The chloride content is lower but it is most constant. It does not exceed 0.74 meq/100 g.

The total amount of water-soluble salts (anion sum) in loess does not exceed 6.50 meq/100 g. In 85 – 90% of the cases it is between 2 ad 4 meq/100 g. The pH value fluctuates from 7.1 to 9.6 (pH about 8 for our samples), i.e. it varies within a relatively narrow interval, so that loess exhibits neutral to slightly alkaline reaction.

Sorption capacity. This parameter shows what part of the cations in the soil could be exchanged by an equivalent quantity of other cations introduced from the outside. Its value depends on the dispersity, on the fine fraction mineral composition and especially on pH of the medium. The sorption capacity of a soil – a host medium for a RAW repository, is one of the important criteria for evaluating its qualities as a natural barrier for radionuclide retention.

It is known that under other equal conditions the bentonite (montmorillonite) clays exhibit the highest exchange capacity (80-100 meq/100 g) compared to other clays due to their specific structure. It is considerably lower in the case of illite (40-20 meq/100 g) and goes down to 15 and 3 meq/100 g. The organic substances increase strongly the exchange capacity. The exchanging complex consists of Ca^{2+}; Mg^{2+}; Na^+ and K^+. The sorption capacity of loess changes from 5 to 15 meq/100 g. It is closer to the lower limit (Table 2) – 5.88 meq/100 g for the sandy loess sample investigated by us and 6.20 meq/100 g for the silty loess sample.

The sorption capacity increases to the south in the fossil soils too, which is connected with the higher clay content and with its higher degree of montmorillonitization. The sandy and silty loess possess lower exchange capacity compared to the clayey varieties. The determined sorption capacity is typical for the loess in the studied region. Its value is not high, which means that the natural barriers should be combined with artificial ones in order to minimise the radionuclide migration in the geosphere.

Structure of loess

The structure of loess is formed under the influence of its specific aeolian origin, its mineral and chemical composition, the dry climate during its deposition as well as of the diagenetic processes. As a result of all this loess had acquired insufficiently compacted and porous structure, its most important specific feature being the instability of structural bonds under the action of water.

The basic building elements of the loess structure are the elementary particles (mono-grains) and the aggregates, connected with inter-particulate and inter-aggregate bonds of colloid, condensation and crystallisation type. According to Minkov (1968) the structure can be granular, aggregate and transitional (aggregate-granular and granular-aggregate) depending on the degree of aggregation, the quantitative criterion being the percent composition of the fraction <0.005 mm, obtained after 20 minutes of mixing.

The loess structure can be represented in other ways too – for example, by the two-dimensional structural model of loess based on a simplified version of the "Monte Carlo" method (Smalley, 1978) or the so-called "masonry" model, representing the building elements of loess in the form of differently arranged bricks (Shen & Hu, 2000). The described structural models are based on various principles and rather complement than exclude each other.

In the present study the structural model of Minkov has been accepted as most suitable for the aspect of consideration. The loess in the investigated region has a granular and transitional structure with the following characteristics:

The granular structure is characterised by deficiency of clayey substance and as a result the latter realises mainly single contacts between the grains of the sandy and silty fraction. The colloidal clay (<0.001 mm) is observed mainly in the form of incrustation of clayey-carbonate substance on the quartz grain surface. In some cases these incrustations (micro patina) serve as a "binder" between the single mono-grains. The total amount of aggregates is small and the predominating ones possess bonds of the condensation and crystallisation type. The granular structure is weak because of the small number and brittleness of the bonds (mainly of carbonate substance) and it collapses spontaneously under conditions of water saturation.

The aggregate-granular and granular-aggregate structure, called generally "transitional" types of structure, are characterised by the fact that the particles of the sandy and silty fraction are not in contact between themselves but "float" in the rest of the silty mass still looking as single grains. The percent of the clayey substance increases in comparison with the former structural type (remaining however still low as an absolute value, especially for the aggregate-granular type) and the number of contacts is increased. The relative number of the structural bonds is low because this structure is characterised by high super-porosity. The aggregates with coagulation and crypto-crystallisation type of bonds are predominating. The structure possesses (similarly to the former type) a low strength in the water-saturated state. Another specific feature of this structure is the regular (isomorphic) distribution of the building elements and structural bonds.

DEPENDENCE OF THE GEOTECHNICAL PROPERTIES ON THE COMPOSITION AND STRUCTURE

A clearly expressed relationship exists between the geotechnical properties of loess and its composition and structure. The main unfavourable property of loess – its collapsibility, is due to the insufficient compaction of the above-described structures and the presence of weak unstable to water action bonds between the sandy and silty grains. These specific features determine the great differences in the strength and deformation properties of loess in the natural and in the water-saturated state.

Average values of the physical and mechanical parameters of loess in the NPP zone (Table 3) are presented here. The loess varieties exhibit low dry density (ρ_d=1.39 – 1.42 g/cm^3) due to their not compacted structure.

The angle of internal friction of the different varieties changes from 27^0 to 30^0, the cohesion – from 10 to 15 kPa, and the modulus of compressibility determined by a stamp is between 14 and 16 MPa for the loess in the natural state. However, this modulus can be reduced to 5 MPa after additional moistening due to weakening of the structural bonds.

From the viewpoint of the considered task, the volume of macropores is of greatest interest. Under vertical loading of 0.3 MPa, which is close to the load transferred to the base of a RAW repository, the volume changes from 4.5 to 6.5%. In some cases the values of this coefficient can exceed considerably the values shown in Table 3. For example, they can exceed 10% for typical loess. This means that the collapse of a massif with a thickness of 10 m can be more than 100 cm. Such big collapse has been observed in irrigation systems to the south of the NPP. This proves categorically that the RAW repository in loess soils cannot be built without preliminary improvement of the soil base. The whole equipment of the Kozloduy NPP has been built on loess after such improvement (Minkov and Evstatiev, 1975), so that significant experience has been accumulated in this respect.

The natural loess represents an unsaturated medium with a low degree of water saturation Sr = 0.30-0.55. This fact presents a number of advantages with respect of RAW disposal – the velocity of radionuclide migration is much lower in an unsaturated medium than in a saturated one. In this connection, the existence of three horizons in the loess massif according to the water content and the moisture fluctuations should be mentioned (Minkov, 1968):

- upper ("impulse horizon") – with a thickness from 3 to 5 m, whose water content is influenced by the seasonal climatic changes;
- medium ("dead horizon") – with a thickness from 5 to 30 m, where mainly century-old moisture fluctuations take place;
- lower ("zone of capillary rising") – connected with the aquifer in the gravel embedded in the base of the loess complex. The height of the capillary rising reaches up to 2.5 m.

The water movement in the two upper horizons proceeds by the moisture movement from thicker to thinner water envelopes, mainly in descending direction. Hence, here the filtration in the sense of Darcy cannot be considered, although the laboratory investigations are carried out with water saturated samples.

Table 3. Average values of physical and mechanical indices of the loess in the Kozloduy NPP zone (by Antonov, 2002)

Loess variety	Solid density	Moisture content	Dry density	Plasticity index	Degree of water saturation	Angle of internal friction	Cohe-sion	Modulus of compressibility	Volume of the macro-pores
	ρ_s [g/cm^3]	W [%]	ρ_d [g/cm^3]	I_P [%]	S_r [-]	φ [degree]	c [MPa]	M [MPa]	% (at 0,3 MPa)
Sandy loess	2,74	11,1	1,42	6,4	0,30	30	0,010	16	4,5
Typical loess	2,72	14,2	1,39	10	0,45	27	0,015	14	6,5

The filtration properties of loess depend on its composition and structure. The sandy loess with its granular structure is more water permeable compared with the typical one due to the increased clayey content and changes to the granular-aggregate structure of the latter. It could be assumed for preliminary assessments that the filtration coefficient in the region of an eventual site near the Kozloduy NPP varies between 1.7x10^{-5} and 5.0x10^{-6} m/s.

CONCLUSIONS

Stable minerals participate in the mineral composition of loess, which would not be subjected to unfavourable diagenetic changes in the course of a several centuries long period – the operation term of the repositories for low and intermediate level radioactive waste disposal. The water-soluble chemical compounds represent a small percent of the total mass and it is not expected that they would exhibit aggressiveness towards the building materials.

The existence of the described type of salinity, the displayed slightly alkaline reaction as well as the low humus content are good prerequisites for the effective interaction between the solid (mineral) phase of loess with the strengthening agents. No corrosion processes in concrete and the other building materials of the eventual repository could take place due to the alkaline medium.

Loess possesses a certain sorption capacity, which is favourable from the point of view of its role as a protective barrier but it is necessary to investigate its sorption with respect to the radioactive isotopes expected to be released by the eventual RAW repository.

The unfavourable specific feature of the loess structure is its collapsibility and high degree of subsidence. However, this feature is prone to control and management due to the granulometric, mineral and chemical composition of loess in the region of Kozloduy.

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