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E.Ya.Kipko, Yu.A.Polozov, Yu.N.Spichak, A.V.Popov

P/A Spetstamgonazhgeologia (STG),349240, Antratsit7d Petrovski St.,Lugansk Region,Ukraine

ABSTRACT

Mining and civil engineering projects in the past have frequently required systems to manage the hydrological consequences of their construction. Mostly these systems were dictated by engineering requirement and involved minimal responses such as lining to reduce leakage or inflow interception or collection followed by diversion, often to waste.

Economic and environmental considerations now supplement engineering requirements such that leakage and inflow prevention must be achieved. Historically such prevention has only been achievable rather uncertainly and/or at high cost by using cement or chemical grouts and liners. This paper introduces the technology involved in formulating and predicting the properties of cost effective stabilized clay sealants.

The prerequisite for achieving successful permeability minimization lies in the purposeful control of the complex physical and chemical stresses which may apply to the sealants in the heterogeneous systems where they are applied. The paper addresses the basic relationships which define structure formation kinetics in stabilized clay sealants, and their dependence upon clay mineralogy. In addition, the integrity of stabilized clay sealant properties against chemical aggression and temperature are examined. Finally, the principal used in formulating stabilized clay sealant hydrological, geotechnical and chemical properties are presented.

INTRODUCTION

Mining and civil engineering structures have suffered from, or resulted in, water inflows variously into or out of their confines. Where this has caused problems, they have been overcome by a combination of systems involving either collection or diversion (e.g.: mine dewatering by sump

INTERNATIONAL MINE WATER ASSOCIATION

E.YA.KIPKO, YU.A.POLOZOV, YU.N.SPICHAK & A.V.POPOV

pumping, channel leakage collection by tile drains), or by introducing sealing system (e.g.: hydrostatic liners in shafts; in-situ abutment grouting around dams, or cement lining or piping along channels).

Not infrequently in mining, and occasionally in or after dam construction, water flows or the probability of water flows may occur which either present a considerable economic penalty or which may threaten the integrity or economic viability of the project (Hancock 1986, Black et. al. 1982, Lyall et;;. al. 1986). These problems have generally been addressed by in-situ placement of sealants - usually cement slurries or chemical polymers.

Except where very thorough drilling and in-situ investigations has been carried out, (for example dam abutments (Alvarez et al. 1985), the operations are uncertain in outcome and fraught with difficulties. In particular, post grouting of fractures which are already contributing flows may prove difficult to impossible with cement slurries due to dilution (Camberfort 1986).

In all cases cement and chemical sealants are expensive, with the greatest expense deriving from the need to incorporate supplemental injection operations to ensure an adequate result (cover) is achieved.

In channel sealing, costs may escalate as a result of the difficulty of the placing an even liner with sufficient structural integrity to have a long life. Equally, the rigidity of cement grouts or liners have lead to failures after set up as geotechnical and other stresses develop as a results of the grouting or sealing; or as a result of continued mining (e.g.: subsidence ref).

Stabilized clay sealants can be used in place of cement or chemical sealants in many instances where either high sealant strength is not required or can be achieved from within the supporting geotechnical mass. Stabilized clay sealants can be formulated from nearly any natural clay to have properties which permit ease of placement and predictable gel properties adequate to ensure that after-movements are minimized permanently to levels which are acceptable. Finally, because they set to a gel, they can sustain a reasonable amount of shear without loss of hydrological or chemical integrity.

COMPOSITION AND PROPERTIES OF STABILIZED CLAY SEALANTS

Stabilized clay sealants comprise systems incorporating clay materials, binding agents, structure forming reagents with a mixing liquid. An important feature of these sealants is the low content of binding agent. This usually does not exceed 15% of the total quantity of constituents (/l, 2/).

Basic properties of stabilized clay sealants are:

- gravitational-density and spreadability,
- · rheological static and dynamic shear strength, structural-viscosity and
- physico/mechanical plastic strength and structure formation kinetics.

INTERNATIONAL MINE WATER ASSOCIATION

STABILIZED CLAY SEALANTS FOR MINING AND HYDRAULIC STRUCTURES

Density and spreadability properties are controlled by the formulation and are controlled and monitored during sealant mixing. Rheological characteristics are similarly determined being measured with the aid of rotational viscosimeters. Physico-mechanical properties are tested by cone penetrometers. Testing for other parameters may be performed on site or in specialized laboratories, depending upon the magnitude of the operation being undertaken.

Analytical and laboratory studies, industrial trials and experience indicate that the main criterion for efficiency are the sealing (permeability) characteristics of the stabilized clay developed in-situ. Efficiency does not relate to mechanical strength. Studies have shown (<u>Reference required</u>) that the strength of sealant material need only be that which is required to withstand the applied hydrostatic pressure. This condition can generally be met at values of plastic (gel) strength on set up not exceeding 0.15-0.20 MPa provided that there is geotechnical strength in the surrounding rock or soil formations.

STRUCTURE FORMATION PROCESSES IN STABILISED CLAYS

Stabilized clay sealants are essentially derived from clay minerals moved intimately with water, and with cement or fly ash as the structure forming reagents. X ray diffraction, electron microscopy and chemical analyses have shown that the sealants upon gelling form a micro-heterogeneous matrix of different mineral components which encapsulate the hydrated clay molecules and take up the space between the clay particles.

The interaction of clay particles with water is inseparately linked with surface dissociation of clay minerals and the formation of ion diffusion layers near the surface of the clay particles. All interactions of clay with water are confined to bonds of a <u>physical</u> nature. On introducing cement into a clay slurry, the hermodynamic balance of the system is sharply disturbed. The basic difference between the cement and the clay particles is that the cement particles are active agents capable of <u>chemical</u> interaction with water. In the course of this interaction three typical reactions can occur:

$$A + H_20 = A'H_20 + A''H_20$$

 $A + H_20 = AH_2O$
 $A + B + H_20 = ABH_20$

Within these reactions lies the basis for structure formation and the generation of stabilized clay sealant characteristics.

The initiation of structure formation is resultant from effects on the thickness of the hydration shells which surround the clay particles in the microheterogenous system, as well as on the emergence of new minerals resultant from hydrolysis reactions involving the binding agent. The particles of new minerals emerging during hydration of cement are characterized by high dispersion and a high specific surface (200-400 m²/g). The types of new minerals formed and their chemical composition

INTERNATIONAL MINE WATER ASSOCIATION

E.YA.KIPKO, YU.A.POLOZOV, YU.N.SPICHAK & A.V.POPOV

contributes strongly to the thermodynamic stability of the resultant sealant system. Thus, manipulation of these is undertaken to vary the sealant properties.

Structure formation in stabilized clay systems has a specific character. On the one hand new minerals capable of reacting with water must emerge in the system, otherwise no growth of strength is observed. On the other hand, these new minerals do not initially form a continuous crystalline lattice, because of the high level of water saturation and the presence of clay particles of differing crystallo-chemical structure and water interaction properties.

Over the total setting time, stabilized clay cement systems are characterized by plasticity and have Bingham-Schwedov viscous plastic body properties. In summary, the growth of strength within stabilized clay systems is inseparably linked to the formation of a mixed crystalline-coagulation structure; therefore, the prerequisites for the setting and growth of plastic strength in a stabilized clay sealant system related to the emergence of hydration compounds derived from the binding agent. These eventually chemically immobilize the water and clay particles.

The generation of structure forming minerals is a spontaneous process resultant from the chemical thermodynamics operating between the water, the clay and the binding agents present. However, because of the initial dispersion and heterogeneity of the slurry, the structures which eventually generate to give the sealant its operational properties, are only achieved when thermodynamic equilibrium is established within the overall medium. This occurs when balance is achieved mechanically, thermally and chemically or stated simply, when macroscopic movement ceases, and when chemical and thermal convective gradients balance out within the sealant and with the containing environment.

The properties exhibited by the sealant under final thermodynamic equilibrium are those which guarantee the sealant its long term mechanical, hydrological and physical stability.

Initially, the thermodynamic balance of the system is continuously disturbed mechanically by mixing and flow stresses. This leads to constant dissolution and recrystallization of the initial structure forming minerals. This continuously disturbs the chemical equilibrium by altering cationic and anionic balances, resulting in the clays present, dependant upon mineralogy, adsorbing and desorbing cations and varying the availability of water within the slurry for mineral hydrolysis reactions.

Since no continuous crystalline lattice develops between clay particles and the structure forming minerals, substances of various crystalline structure can be used as binding agents (cements, slags, caustic magnesite or dolomite, gypsum). These agents exhibit high water demand upon hydration and generate high specific surface structure forming minerals which can encapsulate hydrated clay molecules chemically in a thermodynamically balanced system.

Employing the use of X-ray diffraction, mass spectrography and gas chromatography techniques the kinetics involved in reaching chemical thermodynamic balance and structure formation is seen to be represented by three phases of varying duration.

Phase 1 lasts up to two hours and involves dissolution of cement particles introduced into the clay slurry. This phase is characterized by a marked decrease in the specific electrical resistance of the slurries as hydrolysis releases the binding agents component ions.

INTERNATIONAL MINE WATER ASSOCIATION

STABILIZED CLAY SEALANTS FOR MINING AND HYDRAULIC STRUCTURES

- Phase II lasts between 15 and 20 hours and is associated with intensive hydration of cement particles and the forceful crystallization of new hydrated compounds within the macropores of the slurry. These minerals both take up space and water and ensure a rapid growth of strength and impermeability in the slurry.
- Phase III represents a continuity of Phase II and involves a slow rise in strength and sealing properties as the cement hydration mineral crystalline matrix further generates and links up to totally encapsulate the hydrated clay particles.

In Figure 1a the specific resistance (ρ) of four different clay minerals are represented. These are kaolinite (1) smectite (3), illite (2) and (4). Figure 1b shows the growth in Plastic Strength (Pm).



Figure 1: Kinetics of structure formation process for sealants based on:

- 1 kaolinite,
- 2 illite,
- 3 smectite,
- 4 cement suspension with water-cement ratio of 0.5

215

INTERNATIONAL MINE WATER ASSOCIATION

E.YA.KIPKO, YU.A.POLOZOV, YU.N.SPICHAK & A.V.POPOV



Figure 2: Dependence of kaolinite sealant plastic strength vs mixing time:

l - no mixing,	4 - 1 hour,
2 - 10 min.,	5 - 3 hours,
3 - 30 min.,	6 - 10 hours,
	7 - 15 hours.



Compacting stress, MPa

Figure 3: Effect of Compaction stress on free water content clay, $\gamma = 1.4$ g/cm³ slurry

- 1 applied stress
- 2 stress relief

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Figure 5: Effect of temperature on Plastic Strength at time 1 - kaolinite based sealant

- 2 illite based sealant
- 3 kaolinite and illite based sealant

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E.YA.KIPKO, YU.A.POLOZOV, YU.N.SPICHAK & A.V.POPOV

The duration of each phase and the sealant properties which are generated in each phase can be controlled, in part by selecting clays of different mineralogy and/or in part by changing the chemical or mechanical processes. For example as is shown in Figure 2, intensive mixing in Phase 1 expedites the hydration of the binding agent and increases the strengths achieved in later phases. Significantly, it also alters the strengths achieved relevant to clay mineralogy. This can be seen by comparing Figure 2 and Figure 1b.

Thus, from an experimental viewpoint, it is possible to monitor the course and trends of structure formation by tracking specific resistance and plastic strength, and to experiment with the effects of disturbing chemical and mechanical balance in order to achieve required sealant properties.

SEALING PROPERTIES

The extremely low permeabilities which are achievable within stabilized clay sealants result from the irreversible generation of cement hydration mineral crystals within the interlayer pores of the hydrated clay lattice and the supplementing of the Van de Waal forces acting on the clay hydration layers by chemical bonds. These restrict the mobility of the residual water in the inter molecular layers and causes the mass to react with quasi viscous and thixotropic properties (Non Newtonian).

Studies of compression and decompression characteristics reveal the differences between a simple clay slurry (Figure 3) and a stabilized clay sealant (Figure 4).

Upon compression (Figure 3, line 1), the free water in the clay slurry is expelled only to be reabsorbed in part on decompression with a loss of plastic strength (Figure 3, line 2). The stabilized clay sealant on the other hand loses water on compression (Figure 4, line 1), but because the interlayer pore spaces are then more efficiently blocked and the clay particles more tightly bound there is no water uptake on decompression (Figure 4, line 2) and the plastic strength and sealing capacity is permanently increased.

It is thus apparent that it is the emergence of cement hydration compounds in the macropores of the clay slurry which provides both the required strength of the system and its high sealing (low permeability) properties.

The concentration of binding agent added to a clay slurry increases the concentration of hydration minerals in the clay slurry macropores and increases the strength characteristics of the sealant structure. For example the relationship between one day plastic strength Pm (KPa) for stabilized clay sealants based on kaolin and normal Portland type cements has been found empirically to be

described by the following relationship

Pm = 0.49C + 19

where

218

Pm - plastic strength of 1 day in Kpa C - kg of cement added/m³ of clay slurry where the clay slurry density was 1,2 g/cm³

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The rate of hydration mineral formation and cross bonding which generates strength within the clay sealant is dependant in part upon temperature. This is shown for three different stabilized clay slurries in Figure 5.

Figure 6 shows X ray diffraction wavelength spectra for the three stabilized clays measured after day still stand. It is clear that there are a wide range of structure forming mineral present. The principal plastic strength characteristics at one day however derive from those exhibiting a wave-length of 3.03 Angstrom units $(1 \times 10^{-9} m)$.



Figure 6: X Ray diffraction spectra for stabilized clay sealants (sedimented aggregate) under temperature of: 1 - 20°C, 2 - 40°C, 3 - 60°C

SUMMARY

Stabilized clay sealants can be formulated to meet the demands of virtually any physical or chemical environment. They can be formulated efficiently and cost effectively for any normally occurring clay. The success of their formulation and placement depends upon the thorough evaluation of the physical and chemical environment and of the problems to be confronted in their placement. Given these, the results which can be expected will be predictable and the costs easily determined.

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