DISPOSAL OF URANIUM TAILINGS AS PASTE

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ABSTRACT

Disposal of mine tailings as a paste incorporating Portland cement can provide environmental benefits by reducing contaminant movement. A proposal to use the method to dispose of uranium ore tailings at the Jabiluka mine in northern Australia is being investigated. The mining lease is within the boundaries of Kakadu National Park, a designated World Heritage Area, so particular care is being taken to ensure that groundwater in the park is not deleteriously affected by tailings disposal. No description of a comparable uranium tailings disposal project has been found in existing literature.
This paper discusses paste preparation, paste properties and the benefits of using paste technology. The effects of several factors which control paste properties and may affect subsequent leaching of contaminants from the paste mass are discussed in detail. They include tailings properties, type of cement, tailings water composition and emplacement methods. Chemical and physical aspects related to forming a stable tailings deposit which will immobilise contaminants to the maximum extent are emphasised.

INTRODUCTION

Disposal of mine tailings as a paste after partial dewatering and addition of a small percentage of ordinary Portland cement is being regarded increasingly as good practice in the mining industry. The method has the potential to bind possible groundwater contaminants into a solid mass and thus reduce adverse effects on the environment. The paste can be disposed of in surface or underground repositories. If paste is deposited in worked out stopes, it can be given sufficient strength when cured to support further mining.

There is a current proposal to use the method to dispose of tailings at the Jabiluka uranium mine which is in the early stages of development in the Northern Territory, Australia. The mining company, Energy Resources of Australia (ERA), would prefer to process Jabiluka ore at the existing open pit Ranger mine, some twenty kilometres distant. Tailings would be stored there in the open pit with Ranger tailings. However, problems obtaining approvals from the indigenous people to transport ore through territory under their control have forced the company to consider using a processing facility on the Jabiluka mining lease which is surrounded by, but does not form part of, Kakadu National Park, a World Heritage Area. ERA proposes to store the tailings from a processing plant at Jabiluka as a paste after adding Portland cement.

An earlier proposal involved backfilling stopes with about half the tailings paste and disposing of the remainder in two specially constructed deep open pits in the Kombolgie sandstone formation which overlies the ore-containing schist. However, a preliminary examination of possible sites for the pits indicated deep weathering and relatively high permeability of the sandstone to depths up to 50 m. Consequently ERA revised the tailings disposal plan to replace the pits with specially constructed underground silos.

Current approvals from the Australian Government require tailings in excess of those which can be accommodated in mine workings to be stored in deep underground repositories. Comprehensive investigations of paste properties are being undertaken by ERA, together with environmental studies including an examination of the possible impact of tailings disposal on the groundwater and surface water environment of Kakadu National Park. Regulating authorities have also commissioned studies of the potential impact of the mine and tailings on the park environment.

This paper discusses factors specifically related to paste which incorporates low levels of radioactive elements, in particular uranium and radium, and other potential groundwater contaminants such as magnesium sulfate and manganese. The last two can occur in relatively high concentrations in pore water in tailings as a result of recycling water in the uranium extraction process. Physical and chemical requirements related to paste setting, strength, plasticity and permeability which will minimise contaminant pickup by groundwater will be addressed.

Although ERA's current proposal for disposal of tailings at Jabiluka involves only underground storage of paste, disposal both underground and in open pits is covered in this paper for the sake of generality

A search of the literature and enquiries addressed to specialists involved in mining and hydrogeology have not revealed any case where the paste method has been used in the disposal of tailings produced during the production of uranium oxide from uranium ore. It is hoped that discussion of this paper will provide information on other investigations into the disposal of uranium ore tailings after the addition of cement to form a setting paste.

REVIEW OF PASTE TECHNOLOGY

Paste technology has been used relatively widely in underground backfilling applications because of its economic and practical advantages over hydraulic sandfill and cemented rockfill (Millette et al., 1998). A similar technology has now been developed (and continues to be refined) for treatment and disposal of tailings both underground and at the surface (Brackebusch and Shillabeer, 1998).

Pastes are defined as dense, viscous mixtures of tailings and water which, unlike slurries, do not segregate when allowed to rest (Cincilla et al., 1997). The most important distinguishing characteristic is the grain size distribution of the tailings solids. Empirical data indicate that to form a paste, tailings must contain at least 15% (by weight) of fine solids that pass a 20 µm filter. This fine fraction enables the paste to flow through a pipe without segregating and also increases the water retention capacity because of the ability of the colloidal fraction to retain large volumes of water at their surface. Paste produces a plug flow when transported through a pipeline. Furthermore, because of its non-segregating behaviour, pipe transport is not limited by a "critical" flow velocity, as is the case for slurry transport (Verburg, 1997). Materials without ultrafine particles will not form pastes.

Paste has a low permeability because of the presence of the ultrafine particles which fill voids between the larger particles. Low permeability limits water and air flow through the paste, thus limiting oxidation of sulfides and leaching of metals.

The pressure gradient required for pipeline transportation of paste is much higher than that required for dilute slurries. Much more pumping energy is therefore required to deliver the paste from the mineral processing plant to the tailings deposit. Practical pumping distances range up to three kilometres (Brackebusch and Shillabeer, 1998). In the case of underground mines with surface treatment facilities, gravity can supply most or all of the pressure head required to deliver paste to backfill mine workings or other underground repositories.

Proponents of the paste disposal method claim significant environmental benefits including elimination of surface impoundment of waste liquids, reduction in leachate generation due to high water incorporation capacity, the ability to "engineer" a paste suited to the waste which will enhance environmental benefits, increased rates of filling (rise) compared to slurry disposal and enhanced flexibility in placement practice (Verburg, 1997; Cincilla et al., 1998).

Paste additives

A basic tailings paste consists of tailings and water. Additives, such as Portland cement, are commonly incorporated into tailings pastes to increase strength and durability. Published data for pastes with various percentages of cement addition (Bodi et al., 1996) indicate that uniaxial compressive strengths range from 250 kPa to more than 2,100 kPa for Portland cement percentages of 4 to 16%, with curing times between 7 and 28 days. Cincilla et al. (1997) indicate that cement additions as low as 1% can produce a significant increase in strength. Unconfined compressive strength (UCS) results for 7 day cured full plant tailings yielded UCS values of approximately 70, 150 and 220 kPa for 0, 1 and 3% Portland cement additions.

Choice of additives

Ordinary Portland cement (OPC) has been widely used as a binding agent for tailings pastes and is considered to lead to reduced permeability of the paste, entrapment of contaminants through microencapsulation and generation of neutral to alkaline conditions which generally favour metal contaminant fixation (Verburg, 1997). Other additives, such as fly ash and ground furnace slags, have been used, often in combination with OPC (Chen et al., 1998). These waste products have the advantage of lower cost compared to OPC and alternative binders with even higher water retention capacities than OPC (Sun et al., 1998).

Glasser (1997) notes that the introduction of fly ash and slag does not dramatically affect the paste porosity but does reduce pore connectivity, with the result that well-cured blends may achieve lower permeabilities (< 10^{-12} m/s). This process, known as pore refinement, has been explored by a wide range of methods (Hooton, 1986). It should be recognised that pore refinement in cement and its blends occurs relatively slowly. With good curing, significant decreases in porosity and permeability could be expected over the first 6-12 months after emplacement. However, the extent to which this refinement process would occur in cemented tailings would probably be limited by the low levels of cement addition proposed.

The addition of 1 to 4% by weight of cement is proposed by ERA for paste made from Jabiluka tailings. The percentage would depend on the engineering requirements (particularly strength) of the product. Significant further testing and optimisation of the nature and extent of addition of binding agent(s) will be required prior to final implementation. Some of this work is already being carried out. A 1% addition of cement would appear to be low, particularly in view of the potential for consumption of some of the cement constituents in neutralising the highly acidic process liquors (though lime will be added specifically for this purpose, thereby reducing the demand on cement alkalinity).

De-watering tailings

ERA plans to de-water tailings using a belt filter (ERA, 1998). The solids content achievable through such a process is uncertain, but a "toothpaste consistency" with solids content in the 70-85% range would appear desirable (Brackebusch and Shillabeer, 1998). Some problems can be envisaged in achieving such a solids content, particularly given the presence of significant concentrations of iron oxyhydroxide and siliceous material that will undoubtedly form on neutralisation of the tailings through lime addition. These materials will occur in polymeric, gelatinous form and may lead to significant fouling of the belt filter through formation of a relatively impervious cake.

Significant effort in optimising de-watering procedures is considered essential. Aspects which should be considered are: the pH dependence of belt filter de-watering efficiency; methods to prevent belt filter fouling by generated gelatinous materials (eg through selective use of synthetic polymeric conditioning agents, electro-osmotic enhancement of belt filtration, etc); alternative de-watering methods; the possibility of "staging" the de-watering process with subsequent recombination of the solid phases.

Incorporation of additives into the paste

Effective mixing of cement and de-watered tailings paste will be necessary to ensure generation of a homogeneous product. ERA (1998) mention that a "repulping" of de-watered filter cake and cement will be used to generate a cemented product. More detailed consideration of the mode of mixing may be necessary with consideration given to high-shear colloidal mixers (Reschke, 1998) and other related approaches.

Mode of action of added cement

The mode of action of OPC in the "curing" of tailings paste is considered to be identical to that in normal concrete. That is, hydration of the major components of OPC, namely tricalcium silicate ($3CaO \cdot SiO_2$ or C_3S), dicalcium silicate ($2CaO \cdot SiO_2$ or C_2S), tricalcium aluminate ($3CaO \cdot Al_2O_3$ or C_3A) and tetracalcium aluminoferrate ($4CaO \cdot Al_2O_3 \cdot Fe_2O_3$ or C_4AF) leads to formation of a strong, interconnected solid mass which would be expected to have low permeability, particularly given the relatively fine nature of the constituent solids (Neville and Brooks, 1987). It should be noted however that the presence of significant amounts of high water retention solids, particularly the colloidal iron, manganese and silicon oxides, could result in a modification of the established curing process.

Impact of tailings water composition on cement behaviour

A significant factor which must be addressed is the likely impact of tailings water composition on the strength and porosity of the resultant cemented mass. Both sulfate and magnesium are expected to be present at relatively high concentrations in the tailings water yet both are recognised to retard the setting time and reduce the strength of normal concrete (Neville and Brooks, 1987; Kumar and Rao, 1994). Concerns related to the effect of sulfate on the long term integrity of cemented tailings paste have been expressed by Ouellet et al. (1998).

Cements which are resistant to sulfate attack are commercially available. Type SR cement is reported to be able to withstand sulfate attack at concentrations up to 5% when expressed as SO_3 (approximately 50,000 mg/l) though this relates to the ability of concrete to withstand "external" attack. The rate of deterioration might be expected to be somewhat more rapid when the sulfate is present in the mix water (as it will be in the case of tailings paste).

While precise concentrations of ions in the Jabiluka tailings are uncertain, the major ion composition of process liquors from the nearby Ranger mine provide a guide to what might be expected at Jabiluka (Ranger and Jabiluka ore occur in the same form in the same host rock formation and similar processing is envisaged). Sulfate concentrations approaching 50,000 mg/l are present in process waters and similar concentrations would be expected in tailings waters prior to neutralisation. Addition of lime will result in a pH increase (a pH of 5 is proposed for Jabiluka) and some gypsum ($CaSO_4$) precipitation would be expected. Sulfate concentrations in Ranger tailings waters are typically of the order of 20,000 mg/l. On this basis, it could be expected that more than half of the sulfate would be removed from solution at Jabiluka, though precipitation may result in calcium sulfate accumulation in the solid phase to which cement is being added.

Kumar and Rao (1994) report that the deleterious effects of sulfate peak at 3000 - 4000 mg/l sulfate after which the impact on setting time and strength is not as severe. Indeed, the loss in compressive strength due to the presence of sulfate does not appear to exceed 25% of that observed in the absence of sulfate. While such reductions in concrete strength might be considered critical in the construction industry, the magnitude of such changes would not appear sufficient to seriously limit the use of cement binder for tailings paste, although such effects do flag the need to comprehensively examine the impact of tailings water chemistry on cemented paste behaviour.

While detailed investigation of the effect of tailings water composition on the strength and integrity of the resultant "cemented tailings" must be implemented, consideration should also be given to other means of overcoming this problem. Options include minimisation of the use of sulfate in the ore processing and removal of sulfate (and possibly magnesium) from the tailings water prior to paste formation and cement addition.

The first option is non-trivial and would require a re-assessment of the process used for extracting uranium. The second option is also problematic in that contaminants would need to be removed prior to de-watering and would need to be segregated in some way from other waste solids (which are to be disposed of in paste form). However, neither option should be discounted. The issue of treatment of tailings water prior to reuse in the process (a necessity in the case of Jabiluka because of the zero surface water release policy) demands particular attention from the wider perspective of avoiding problems in the process due to the re-use of waters containing significant levels of contaminants. Not only might scale formation due to precipitation of calcium and/or magnesium solids create problems, but the presence of gelatinous iron and silica polymeric materials could well cause serious problems in solvent extraction and solid-liquid separation steps. Removal of at least a portion of these materials would seem essential given the concentration build-up that would be expected given the requirement of zero contaminant release.

Given the apparent detrimental effect of sulfate on cement integrity, it is interesting to note that recent work has shown that calcined gypsum can be used in place of OPC as a tailings paste binder. Thus, Petrolito et al. (1998) have shown that compressive strengths equivalent to that achieved with OPC can be obtained through use of 2.5 to 4 times the corresponding amount of cement. Given the abundance of calcium sulfate in the tailings, use of an alternative binder of this form has considerable attraction. Obviously, to achieve the binding properties required, calcination would be necessary. Such a process would require the separation of precipitated gypsum from the tailings solids to enable calcining but the returns achievable may justify the considerable development work that would appear necessary to facilitate such an approach.

Emplacement and curing

Paste could be placed in and cured in de-watered repositories or under water.

In the case of disposal in open pits, sub-aqueous deposition would be the most likely since special provision would have to be made to pump rainwater or any liquor bleeding from the paste away to some liquid storage facility. Underground mine workings and underground silos could be backfilled under water, or in the dry if appropriate drainage and pumping were maintained.

Free fall of the paste either above or below a water surface would incur the risk of segregation. This could lead to both chemical and physical inhomogeneity of the deposited paste. The former could result in zones of concentration with increased solubility and leaching of contaminants while the latter could result in more permeable zones of coarse material which would allow groundwater easier passage through the tailings deposit. Both effects would be deleterious in relation to the movement of contaminants from the site. Use of a movable tremie pipe to discharge paste below the surface of previously deposited material and distribute it evenly would be advantageous.

Uneven settlement and shrinkage of the tailings may be sufficient to cause cracking of the tailings mass and thus higher permeability. This is more likely to occur in backfilled mine workings than in the vertical cylindrical silos proposed for Jabiluka because of the irregular shape of the former which will cause stress concentrations during curing. This potential problem needs to be addressed when strength and deformability data are available from laboratory testing.

Another potential problem in the case of backfilling mine workings is leaving voids, particularly at roof level, to form channels through which groundwater could flow freely. These must be avoided to restrict contaminant movement.

SUITABILITY OF JABILUKA ORE FOR PASTE FORMATION

Golder Associates made an assessment of the paste forming potential of the Jabiluka ore using diamond drill core (Golder Associates, 1997). The core was processed in a laboratory to form tailings which would represent those to be produced by the proposed mine. Over 30% of the tailings was found to be less than 20 μm in size, indicating that the tailings would be suitable for paste formation. The results of rheological tests indicated that the properties of trial pastes made from tailings and water (i.e. without addition of cement) were "excellent", with low yield stress and good water retention. The little water that bled from the paste would probably be taken up by the cement which will be added to the tailings under current proposals.

ERA has commissioned consultants to perform a range of tests on pastes containing Portland cement. The results will be available in the near future.

CONTAMINANT MOBILITY

From the environmental viewpoint, the most important requirement of tailings disposal is minimisation of movement of contaminants from the site. It is assumed that erosion of the tailings by the forces of nature will be prevented for an appropriate period by burial at a sufficient depth under a cover which will remain in place. However, even though the tailings are physically immobilised, contaminants can still move from the site in gaseous form or dissolved in groundwater.

Radon gas produced by the radioactive decay of radium is a potential atmospheric contaminant. Its movement is reduced, and dispersion improved, by low permeability of the tailings mass and cover.

Prevention of leaching of contaminants from the tailings mass can be considered as the primary barrier to groundwater contamination. Low permeability of the surrounding rock and adsorption of contaminants such as uranium within the rock provide a secondary barrier. The distances contaminants will move, and concentrations of contaminants in groundwater near the tailings repositories, will depend on the efficacy of both of these barriers and the magnitudes of hydraulic gradients in the groundwater flow system.

The potential of paste technology to improve the physical properties of the primary barrier have been discussed above. Possible problems related to chemistry have also been discussed. It is essential that adequate investigation of both physical and chemical properties of pastes affecting stability and leaching be carried out in particular cases. This data must then be used with appropriate groundwater flow and contaminant transport analyses to predict environmental effects.

Effect of cement addition on contaminant leaching

The addition of cement to tailings paste is likely to have an overall positive effect on contaminant immobilisation. The alkaline conditions that are likely to result from lime and cement addition to the tailings is expected to assist in "fixation" of a range of contaminants either through formation of insoluble oxide/hydroxide forms or through enhanced adsorption to solid phases (particularly the high surface area, amorphous iron, manganese and silicon oxides that will form on pH increase). However, uranium may be mobilised under such conditions. The affinity of uranium for carbonate anions is sufficiently strong for U(VI) to be made soluble at pH values above about 8 where the concentration of carbonate in solution is sufficient to dominate the speciation of U(VI) (Waite et al., 1992; Waite et al., 1994). While this issue is worth noting, a number of factors may counter the possible dissolution of U(VI) at high pH. These are:

- A considerable amount of silicate will be present which may stabilise the uranyl ion either in insoluble uranyl silicates or in surface-bound U(VI)-silica ternary complexes,
- The possibility of formation of insoluble magnesium-uranium sulfates, magnesium-uranium carbonates, calcium-uranium carbonates or calcium-magnesium-uranium compounds exists (Wanner and Forest, 1992; Glasser, 1997). Such solids have been identified as apparently controlling uranium solubility in high pH, high sulfate cement pore waters in northern Jordan (Linklater, et al., 1996),
- Much of the deposited tailings may become anoxic and exhibit reducing conditions as a result, in part, of the inability of oxygen to diffuse into the relatively impermeable, sub-aqueous matrix. Under such conditions, U(VI) would be expected to undergo redox transformation to the U(IV) state where it would be expected to form highly insoluble oxides. While considerable variability has been reported in the solubility products of "amorphous" or "hydrated" forms of UO_2, these solids would be expected to be highly insoluble over a relatively wide pH range (Wanner and Forest, 1992).

Approximately 5% of the uranium in the ore will remain in the tailings. This uranium has survived the harsh physical and chemical treatment applied to the ore to extract as much of the uranium as possible. Consequently only a small proportion of the remaining uranium might be easily accessible to alkaline solutions which could possibly mobilise it and carry it into the groundwater.

Long term stability and contaminant containment

With regard to the ability of cemented tailings to immobilise contaminants, the scientific literature is not definitive. While it might be expected that oxyanions of elements such as arsenic, selenium and chromium would exhibit limited sorptive capacity to surface sites of tailings constituents (Dzombak and Morel, 1990; Glasser, 1997), the evidence suggests that these anions are stabilised to relatively low solution concentrations at high pH, presumably as a result of formation of (often ill-defined) mineral phases (de Groot et al., 1989). The possibility of U(VI) mobilisation as a result of the formation of highly soluble uranyl carbonates has also been mentioned but the likely formation of uranyl silicate and/or calcium-uranium precipitates is expected to mitigate such an effect. Notwithstanding these arguments of likely contaminant immobilisation, comprehensive leaching studies of contaminant stability, particularly as a function of pH and major ion tailings water composition, should be undertaken.

Glasser (1997) points out that generation of gases such as CO_2 and CH_4, mainly as a result of microbiological activity, could lead to cracking and physical deterioration of the matrix. Carbon sources for such activity are likely to be limited however and the potentially high pH conditions may limit microbiological activity. Lange et al. (1996) also note that carbonation of cement-solidified hazardous wastes appears to lead to a stronger rather than weaker solid matrix with enhanced metals retaining ability.

A substantial literature exists on use of cement solidification and stabilisation of hazardous materials, including radioactive wastes (see, for example, Connor, 1990; Glasser, 1997). While the proportions of cement being added are typically somewhat higher than those proposed for tailings pastes, many of the issues raised with regard to fixation of tailings contaminants have been considered to some extent. Close attention to this literature should be maintained as comprehensive test programs are implemented.

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