Use of lignite fly ash as an additive in alkaline stabilisation and pasteurisation of wastewater sludge

In this study, the possibility of using lignite fly ash in low doses for reducing the pathogen levels in wastewater sludge was investigated. The results showed that using fly ash alone in doses of 40%, 80% and 120% (on a dry weight basis), did not produce an alkaline environment for an efficient removal of pathogens. However, using fly ash in conjunction with the minimum amount of quicklime may act as an effective way of fecal coliform removal in both alkaline stabilisation and pasteurisation processes. It was shown that using fly ash in doses of 80% and 120% in alkaline stabilisation and pasteurisation processes prevented the pH decays and regrowth of pathogens during 60 days of storage period. The results of the study confirmed that alkaline pasteurisation process produces a product which is more resistant to pH decays and regrowth of fecal coliforms compared to that of alkaline stabilisation. Consequently, the overall results of this study indicated that the minimum lime and fly ash dosages required to generate a Class B biosolid were 10- 15% and 80%, respectively. On the other hand, heating sludge to 50°C prior to the addition of 10-15% quicklime and 80% fly ash followed by further heating to 70˚C and then sustaining at this temperature for 30 minutes were sufficient to generate a Class A biosolid.

Introduction

Combustion of coal produces a variety of waste materials such as fly ash, bottom ash, flue gas desulfurisation waste and coal gasification ash. Fly ash is the mineral residue resulting from the combustion of coal that enters the flue gas stream. It is composed predominantly of fine particles and is either collected in emission control devices or released from the stack. There are basically two methods for the disposal of fly ashes: settling ponds and landfills. The major adverse impacts of ash disposal on terrestrial ecosystems include groundwater contamination, reductions in the growth of crops and changes in elemental composition of crops. Ash disposal in landfills and settling ponds can also influence adjacent aquatic ecosystems

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directly through inputs of ash basin effluent and surface runoff (Carlson & Adriano 1993). Hence, fly ash disposal sites must be well managed in order to protect the local surface and groundwater supplies. This can cause significant economic burden to achieve the necessary water and land management. Therefore, a great deal of research has been conducted to identify and determine the feasibility of utilising these wastes in industrial and agricultural applications.

Fly ash has been successfully used as a mineral admixture in portland cement concrete and asphalt concrete and as an embankment or structural fill material (Krishnamoorthy *et al*. 2002, Prashanth *et al*. 2001, Palomo *et al*. 1999). In addition, fly ash has a vast potential use in agriculture as an agronomic amendment specially, for its physical characteristics which are conducive for plant growth due to the presence of macro and micro nutrients (Wallace *et al*. 1980, Elseewi & Page 1984).

Alkaline stabilisation of wastewater sludge is another utilization area for fly ash. Poon & Boost (1996) used a slightly modified process by adding pulverized fly ash/ quicklime mixture to the sludge. They suggested that the use of pulverized fly ash in conjunction with lime is sufficient to produce a final product with improved handling characteristics and reduced leaching potential. Another study indicated that the wastewater sludge conditioned by fly ash can be disposed off in landfills and used as a soil conditioner (Wang & Viraraghavan 1997).

These studies generally focused on the use of high fly ash dosages. Poon & Boost (1996) added fly ash in the range of 342-410g and quicklime in the range of 41-410g to 590g sludge (13.7% dry solid) on a wet weight basis. Fly ash/sludge proportions (w/w) of Wang & Viraraghavan (1997) were 1:1, 1:3 and 1:9. They proposed that mixing wastewater sludge with alkaline fly ash was a reasonable way for land application without the possibility of leachate contamination and in that way it may be safely used as a cover material for landfill and as a landfill engineering material. However, application of biosolids having high amounts of fly ash in agricultural soils may be limited due to possible negative impacts of fly ash.

Trace element concentrations in fly ash were found to be higher than those in coal and the concentrations of biologically toxic elements such as B, Mo and Se may greatly exceed their background levels in soil (Kalra *et al*. 1996). It is evident that if the treated sludge is used as a fertilizer or as a soil conditioner in agricultural soils, the end-product should not contain excessive amounts of fly ash. Therefore

it is very important to determine and evaluate the effects of using lower doses of fly ash in alkaline stabilisation of wastewater sludge.

The objectives of this study were to examine the possibility of using lignite fly ash in low doses to decrease the pathogen levels in wastewater sludge and to investigate the effects of fly ash in conjunction with quicklime on alkaline stabilisation and alkaline pasteurisation of wastewater sludge. It was also aimed to observe the changes in stabilised sludge mixtures during the storage period of 60 days in order to determine the long term effects of fly ash on pH decays and regrowth of pathogens.

Materials and Methods

Materials

Three wastewater sludge samples which were different in origin, were collected from Pirelli Cable Systems Company, Mauri Yeast Company and Penguen Canned Food Company. Wastewater sludge from Pirelli (Pi-WS) originated from the treatment of domestic wastewater. Wastewater sludges from Mauri and Penguen (Ma-WS and Pe-WS respectively) originated from the treatment of both domestic and industrial wastewaters.

The fly ash used in this study was obtained from Orhaneli Power Station where lignite is used for fuel. Technical grade anhydrous calcium oxide (quicklime 96%) was used together with fly ash in alkaline stabilisation and pasteurisation processes.

General characteristics of fly ash and wastewater sludges are reported in Tables 1 and 2, respectively.

Experimental design

Raw sludge samples were treated in three different methods:

Table 1: General characteristics of Orhaneli Power Station fly ash.

11.80-12.10 pH (1:5, solid:water)	
$(\%, w/w)$ P_2O_5 $0.15 - 0.33$	
$\boldsymbol{\mathcal{U}}$ SiO ₂ 46.80-50.30	
$\boldsymbol{\mathcal{U}}$ $\mathsf{Fe_{2}O_{3}}$ 8.50-11.95	
Al_2O_3 $\boldsymbol{\mathcal{U}}$ 15.18-24.40	
\boldsymbol{u} TiO ₂ $0.85 - 1.10$	
$\boldsymbol{\mathcal{U}}$ MgO 2.99-4.01	
$\boldsymbol{\mathcal{U}}$ CaO 8.99-10.75	
\boldsymbol{u} SO ₃ 3.07-5.98	

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Table 2: General characteristics of wastewater sludge samples used in study.

Treatment 1:

Wastewater sludge was mixed with 40%, 80% and 120% of fly ash on dry weight basis.

Treatment 2:

Fly ash-quicklime mixture was added to wastewater sludge to achieve the minimum EPA criteria for Class B lime stabilisation.

Treatment 3:

Fly ash-quicklime mixture was added to wastewater sludge to increase the pH to 12 and the temperature of the mixture was also increased to 70˚C by using a supplemental heat source.

It was generally preferred to use excessive amounts of quicklime in order to heat the sludge to pasteurisation level. However, the utilisation of this end-product will probably be limited in alkaline soils of Turkey. Furthermore, using excessive amount of quicklime will decrease the fertilizer value of the biosolid due to losses of nitrogen as ammonia (Andreasen 2001). Hence, in this study the minimum amount of quicklime and fly ash were used for meeting pH requirement and the temperature regime was maintained by supplemental heat source.

The proportions of wastewater sludge, fly ash and quicklime tested in this study are given in Table 3. A variable speed laboratory paddle mixer was used to mix wastewater sludge with lignite fly ash and lime. Mixing chamber included electric heating elements bolted to the chamber walls. An insulated jacket around the heating elements directed heat into the chamber. This mixing chamber met the temperature requirement to obtain Class A pathogen reduction. A schematic diagram of the experimental set up is shown in Fig. 1.

Methods

All samples were analysed for pH, dry matter, temperature and fecal coliform prior to treatment and at various steps after treatment. Dry matter contents of raw materials and mixed samples were determined according to the Standard Methods (APHA, AWWA & WEF 1998). Metrohm 704 pH meter with a combined glass electrode was used for pH readings. pH values of the samples were measured using a proportion of 1 g solid to 10 ml distilled water after the equilibrium was reached in 10 minutes (Mc Lean 1982). The readings were corrected for 25[°]C, using the equation published by EPA (USEPA 1999). The temperature profiles of the samples were recorded by a thermocouple and a data logger.

Fecal coliform counts were performed in accordance with the most probable number method given by Standard Methods (APHA, AWWA & WEF 1998). Brilliant Green Bile Broth was used as a growth medium. Inoculated tubes were incubated at 44.5 ± 0.2 °C for 24 ± 2 h. Results were obtained as MPN per 100 ml and converted to MPN per gram dry matter.

Results and Discussion

Fig. 1: A schematic diagram of the experimental set up.

Treatment 1

pH, dry matter content and fecal coliform numbers of sludge-fly ash mixtures are given in Table 4. Dry matter content of the sludge samples increased following the addition of fly ash. Lignite fly ash used in this study contained sufficient alkaline materials to have a pH of approximately 12 (Table 1). However, the alkaline level of the fly ash alone did not produce a highly alkaline environment for the stabilisation of sewage sludge as shown in Table 4. The maximum pH increment after 2 hours of contact was only about 0.8 unit even with 120% fly ash dose. It is clear that applied doses of fly ash were insufficient to create an alkaline environment for removal of pathogens.

Figs. 2, 3 and 4 depict the variations in fecal coliform numbers and pH values of fly ash-sludge mixtures during 60 days of storage for Pe-WS, Ma-WS and Pi-WS, respectively. It can be seen from Fig. 2A that the pH values of Pe-WS mixtures decreased to original pH value of raw

sludge after 15, 30 and 45 days of storage for the mixtures with 40%, 80% and 120% fly ash, respectively. While the fecal coliform numbers of the mixtures with 0%, 40% and 80% fly ash were showing a tendency of increase during 60 days of storage, they generally remained constant for the mixture with 120% fly ash (Fig. 2B).

The pH value of the mixture with 40% fly ash decreased to original pH of the sludge only after 15 days for Ma-WS. However, no noticeable pH change occurred in mixtures with 80% and 120% fly ash during 60 days (Fig. 3A). An increase in fecal coliform numbers in Ma-WS mixture with 40% fly ash occurred probably due to the observed decreases in pH (Fig. 3B).

 It can be seen from Fig. 4A that pH values of all Pi-WS mixtures decreased slightly during the storage period. However, the overall decrease is higher in the mixture with 40% fly ash. The fecal coliform numbers of this mixture slightly increased after 30 days (Fig. 4B). It was concluded that the 80% and 120% fly ash dosages had some preventing properties on both pH decays and regrowth of fecal coliforms in Ma-WS and Pi-WS. For Pe-WS mixtures, the fly ash dosage of 120% was found to be the minimum, which showed preventing effect in terms of pH decays and regrowth of fecal coliforms.

It has been recommended that a pH of 10.5 or above is required for an efficient bacterial inactivation (Bitton 1994). Similarly Boost & Poon (1998) revealed that a pH of 11 or greater was shown to inhibit the growth of all enteric bacterial pathogens in sludge. Poon & Boost (1996) reported that the fly ash used in their study alone did not prevent the fecal contamination efficiently. On the other hand, Wang & Viraraghavan (1997) obtained higher pH values with the addition of higher amounts of alkaline fly ash into sludge and they reported that the fly

Fig. 2: (A):Variations of pH in Pe-WS-ash mixtures during 60 days of storage period, (B): Variations of fecal coliform numbers in Pe-WS-ash $(-\bullet, -\bullet \quad \text{WS}, -\bullet \quad \text{vs}, -\bullet \quad \text{WS} \quad 1, -\bullet \quad -\bullet \quad \text{WS} \quad 2, -\bullet \quad -\bullet \quad \text{ws} \quad 3).$

Fig. 4: (A):Variations of pH in Pi-WS-ash mixtures during 60 days of storage period, (B): Variations of fecal coliform numbers in Pi-WS-ash mixtures during 60 days of storage period (-•, -o- WS, -■-, -□- WS 1, -▲-, -△- WS 2, -▼-, -∇- WS 3).

ash can achieve a minimum percentage kill of 93.5% in microorganism population. The differences in results of this study and others may be explained by some variations in properties of fly ash such as soluble oxides and trace metals contained in fly ash particle matrix as well as the fly ash dosages.

Treatment 2

Class B pathogen reduction by alkaline stabilisation requires addition of sufficient alkaline materials to the sludge to raise the pH above 12 after 2 hours of contact. A preliminary study was conducted for determining the optimum proportions of quicklime using in conjunction with lignite fly ash. The results of the preliminary study showed that the pH of Ma-WS and Pi-WS with 10%, 15% and 20% lime dosages remained above 11.5 over 30 days. However the pH of the sludge samples with 5% lime dosage dropped below 11.5 after 5 days. Therefore, it was decided to use a quicklime dose of 10% in alkaline stabilisation of Ma-WS

and Pi-WS. On the other hand, it has been concluded that a quicklime dose of 15% appeared to be the minimum dose required for Pe-WS. The differences in pH values and the buffering capacity of organic mass in sludge samples probably changed the required quicklime dosage (Andreadakis 1999).

 After determining the optimum doses of quicklime, the sludge samples and the alkaline materials were mixed in an insulated vessel in order to minimise the heat losses. Resulting temperature values, pH, dry matter content and fecal coliform numbers of alkaline mixtures are given in Table 5.

The resulting temperature levels of the mixtures showed that the mixing of alkaline materials with sludge did not produce a significant amount of heat for temperature elevation. It can be also seen from Table 5 that no decrease was observed in actual temperatures of mixtures with 80% and 120% fly ash in contrast to expected theoretical temperature levels. A higher fly ash content resulted in a

higher recorded temperature.

Table 5 shows that the pH values of the mixtures were all above 12 after 2 hours of contact time and a significant fecal coliform removal was achieved. After 24 hours of contact, the density of bacterial indicators decreased below detection limits in most of the alkaline mixtures even with doses of 10% quicklime and 40% fly ash. In order to be effective, lime stabilisation must achieve a pH of 12 or above for at least 2 hours (Bitton 1996). Westphal and Christensen (1983) showed that fecal coliform reductions at 2 hours averaged more than 5 logs and they proposed that achieving a pH \geq 12 at 2 hours after lime addition is a good measure for the effectiveness of stabilisation processes.

Brewster *et al.* (2002) investigated the disinfection of biosolids with low lime doses and fly ash. They used lime doses of 50g, 100g and 200g per kg of biosolids (dry) along with fly ash doses of 500g, 1000g and 1500g per kg of biosolids (dry). The results of their study indicated that fecal coliform bacteria and reovirus were completely inactivated for doses as low as 100g lime per kg biosolids (dry) or 50g lime +500g fly ash per kg biosolids. Ascaris eggs were also removed by using 100g lime per kg biosolid, which produced a Class A biosolid in terms of pathogen removal. The enhanced removal of pathogens with low doses of alkaline materials was explained by 69 days of storage under anoxic conditions.

Treatment 3

Alkaline stabilisation processes can also produce Class A biosolids with respect to pathogens. The temperature requirement can be achieved by overdosing with quicklime, by using a supplemental heat source or by using a combination of the two (USEPA 2000). In this study it was decided to preheat the sludge cake before the addition of quicklime. The slaking temperature is one of the factors which affects slaking efficiency by influencing specific surface of the calcium hydroxide. When a cool water and lime come in contact a condition called "drowning" takes place. Particles of hydrate formed under "drowning conditions are very coarse and not very reactive (Boynton 1980). Therefore, preheating of the sludge cake probably enhances the hydration reaction which occurs during slaking of quicklime and significantly enhances the exothermic effect of the quicklime. In this study it was decided to heat wastewater sludge samples to 50˚C prior to the addition of alkaline materials. After the addition of alkaline materials, mixtures were heated to 70˚C and sustained at this temperature for 30 minutes. pH, dry matter contents and fecal coliform numbers of alkaline pasteurised mixtures are given in Table 6. The results show that pH values of all pasteurised mixtures were above 12. The dry matter contents of mixtures immediately increased due to the addition of dry substances and loss of water from sludge during the heating period. The pasteurisation criteria of 70˚C for 30 minutes and highly alkaline environment reduced the density of bacterial indicators below detection limits.

Figs. 5, 6 and 7 show the pH values and fecal coliform numbers during 60 days for alkaline stabilised and alkaline pasteurised mixtures of Pe-WS, Ma-WS and Pi-WS, respectively. It can be seen from Figs. 5A and 5C that the pH values remained almost constant during 30 days in both alkaline stabilised and alkaline pasteurised of Pe-WS mixtures. After 30 days, a decrease in pH was observed

Table 5: Temperature profiles, pH, dry matter content and fecal coliform numbers of alkaline stabilised sludges.

for all of the mixtures. However, the pH decay in alkaline pasteurised mixtures was generally lower than that of in alkaline stabilised mixtures. It appears that lower pH decays occur with the higher doses of fly ash. An increase in fecal coliform numbers was observed in alkaline stabilised mixtures during 60 days, especially in mixture with 40% fly ash (Fig. 5B). However, no noticeable fecal coliform regrowth occurred in pasteurised mixtures in the end of 45 days, even with 40% fly ash dose (Fig. 5D). It appears that heating of sludge to 70˚C produced a mixture which was more resistant to pH decay and regrowth of fecal coliforms. Fig. 6A indicates that a decrease occurred in the pH values of all alkaline stabilised Ma-WS mixtures. A sudden pH decay was observed in the mixtures with 40% and 80% fly ash after 30 days. On the other hand, there was no sudden decrease in pH values of alkaline pasteurised mixtures (Fig. 6C). The fecal coliform numbers of the stabilised mixture with 40% fly ash increased significantly after a 15 days period (Fig. 6B). Besides, a slight increase was also observed in alkaline pasteurised mixtures after 30 days, even in mixture with 40% fly ash. The variations of pH and fecal coliform numbers of alkaline stabilised and pasteurised Pi-WS mixtures were similar to those of Ma-WS mixtures (Figs. 7A, 7B, 7C and 7D).

The results of the overall fecal coliform log reductions in alkaline stabilised and alkaline pasteurised sludge samples after 60 days of storage period are given in Table 7. The overall fecal coliform log removal values show that alkaline pasteurisation process achieved higher log reductions in all sludge samples. Table 7 also indicates that increasing amount of fly ash resulted in higher log removals.

Correlation analyses were also carried out between pH values and fecal coliform numbers for all sludge mixtures and results are presented in Table 8. Correlation coefficients appear to be all above –0.9 and are significant at a confidence level of 0.01. These high negative correlation coefficients indicate a strong relationship between pH and fecal coliform numbers in sludge. Decreasing pH levels are inclined to produce increasing bacterial numbers.

Conclusions

When fly ash was used on its own as an additive of sludges, it didn't produce a highly alkaline environment for fecal coliform removal. The present study suggested that using lignite fly ash in conjunction with quicklime is an efficient way of pathogen reduction. In mixtures with 80% and 120% fly ash pH decay or regrowth of fecal coliform didn't occur possibly due to the stabilisation effect of fly ash. However, the pH decay and regrowth of fecal coliforms generally occurred in mixtures with 40% fly ash.

On the other hand, it was proved that alkaline pasteurisation process produced a mixture more resistant to pH decays and regrowth of fecal coliforms during a stor-

Fig. 5. (A): Variations of pH in alkaline stabilised mixtures of Pe-WS during 60 days of storage period, (B): Variations of fecal coliform numbers in alkaline stabilised mixtures of Pe-WS during 60 days of storage period, (C): Variations of pH in alkaline pasteurised mixtures of Pe-WS during 60 days of storage period, (D): Variations of fecal coliform numbers in alkaline pasteurised mixtures of Pe-WS during 60 days of storage period (-●-, -o- WS, -■-, -**□**- WS 7, -▲-, -∆- WS 8, -▼-, -∇- WS 9).

Fig. 6: (A): Variations of pH in alkaline stabilised mixtures of Ma-WS during 60 days of storage period, (B): Variations of fecal coliform numbers in alkaline stabilised mixtures of Ma-WS during 60 days of storage period, (C): Variations of pH in alkaline pasteurised mixtures of Ma-WS during 60 days of storage period, (D): Variations of fecal coliform numbers in alkaline pasteurised mixtures of Ma-WS during 60 days of storage period (- -, - - WS, - -, - - WS 4, - -, -Δ- WS 5, - -, -∇- WS 6).

Fig. 7: (A): Variations of pH in alkaline stabilised mixtures of Pi-WS during 60 days of storage period, (B): Variations of fecal coliform numbers in alkaline stabilised mixtures of Pi-WS during 60 days of storage period, (C): Variations of pH in alkaline pasteurised mixtures of Pi-WS during 60 days of storage period, (D): Variations of fecal coliform numbers in alkaline pasteurised mixtures of Pi-WS during 60 days of storage period $(- \bullet, - \bullet - \mathsf{WS}, - \bullet - \bullet - \mathsf{WS}, - \bullet - \bullet - \mathsf{WS} \cdot \mathsf{A}, - \bullet - \bullet - \mathsf{WS} \cdot \mathsf{S}, - \bullet - \bullet - \bullet - \mathsf{WS} \cdot \mathsf{S}).$

Table 7: The overall fecal coliform log removals of alkaline mixtures after 60 days of storage period.

Table 8: The correlation between pH and fecal coliform numbers in alkaline stabilised and pasteurised mixtures.

age period of 60 days. This situation was probably due to the effect of temperature on the slaking process of calcium oxide. Process of heating sludge before the addition of quicklime and fly ash resulted in calcium hydroxide with finer particle size and greater specific surface area. This may have prevented the probable pH decays as a result of enhanced contact between sludge solids and lime.

Correlation analyses indicated a strong negative relationship between pH and fecal coliform numbers in all stabilised and pasteurised mixtures.

The overall results of this study indicated that the minimum quicklime and fly ash dosages required to generate Class B biosolid were 10-15% and 80%, respectively. The final Class B biosolid product can be stored at least for 60 days with minimum pH decay and without pathogen regrowth. On the other hand, heating sludge to 50˚C prior to the addition of 10-15 % quicklime and 80% fly ash followed by further heating to 70˚C and then sustaining at this temperature for 30 minutes were sufficient to generate a Class A biosolid. However additional studies must be done for determining the optimum preheating temperature.

Process of using lime to simply raise the pH above 12 in combination with electric energy to generate heat appear to have some significant advantages over most other lime

stabilisation processes. This process produces a Class A biosolid in fully controlled conditions. The use of an insulated mixing and heating chamber will capture the hydration heat in the process. In lime stabilisation processes (adding CaO only or adding CaO and fly ash) more lime is required in order to meet the temperature requirement. Using excessive amounts of lime will certainly reduce the fertilizer value of the end product and limit the application areas. On the other hand, using fly ash in conjunction with quicklime reduced the amount of quicklime required for pH elevation. Using fly ash as an additive will also enrich the micronutrient content of the biosolid product.

When a treatment plant markets the sludge product as a fertilizer, it is evident that addition of fly ash alone is not an adequate process. The treatment plant should choose the most suitable process (addition of lime and fly ash with or without supplemental electric heat) according to desired disinfection level of the end product. If the purpose is to obtain and market a Class B biosolid, addition of lime and fly ash without supplemental electric heat will be sufficient. However using Class B biosolids in Turkey may cause some environmental problems due to lack of standards set by the environmental authorities. Obtaining a Class A biosolid by the addition of lime and fly ash with supplemental electric heat will be a safer process in terms of agricultural applications. Unlike microbiological properties, no significant differences are expected in chemical properties of biosolids obtained from Treatment 2 and Treatment 3. However, before suggesting the beneficial biosolid to farmers, additional studies should be carried out for determining the effects of heating to 70˚C on the amount and composition of nutrients contained in sludge.

Alkaline stabilisation and pasteurisation processes may be performed in a wastewater treatment plant with uncomplicated facilities and personnel with basic skills. The equipment necessary for alkaline stabilisation and pasteurisation processes is relatively easy to install and operate. Typical equipment includes a wastewater solids feed/conveyance mechanism, lime silo, fly ash silo, alkaline material transfer conveyor and a mixer. Alkaline pasteurisation with supplemental heat source requires an insulated mixing and heating chamber instead of a simple mixer. Both processes can be operated relatively easily. Apparently, operational

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cost of alkaline pasteurisation process will be higher due to heating requirement. However it produces a Class A biosolid with respect to pathogen content.

Finally this study indicates that alkaline fly ash of Orhaneli Thermic Power Plant has the potential of reuse as an additive with respect to pathogen reduction in both alkaline stabilisation and pasteurisation processes. However, before estimating the detailed cost of the processes and evaluating the applicability of the treated sludge on agricultural soils, the fertiliser value of the product must be determined as well as the impact of the application on soils and crops.

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