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Pollution removal from leachate using bottom ash-bentonite-zeolite liner

Ayse Sandikci Mollamahmutoglu, Gurdal Kanat, F. Ilter Turkdogan*

Department of Environmental Engineering, Faculty of Civil Engineering, Yildiz Technical University, 34220, Davutpasa, Esenler, Istanbul, Turkey, Tel. +90-212-3835393, Fax +90-212-3835358, email: ilter@yildiz.edu.tr (F.I. Turkdogan)

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ABSTRACT

There is a crucial need for the development of a landfill liner that can effectively prevent the transport of contaminants in solid waste landfills. The search for strongly adsorbing materials with high treatment efficiency is still continuing. In this study, removal of organics (COD, total-P and TKN parameters) and heavy metals were investigated in the form of a liner formed by mixing the bottom ash and zeolite with bentonite. The various mixture ratios were prepared and studied in order to provide optimum value of impermeability coefficient and heavy metal/organic pollution retention efficiency. The lowest permeability was found to be provided by 30% ratio in bentonite. The overall rate of heavy metal removal from the leachate was up to 98%, while COD, TSS, total-P and TKN were removed in the rates of 98, 85–97, 83–96 and 96–98% respectively. Higher efficiency was found particularly for Zn, Pb and Ni removal. In the study, moreover, cost evaluation was also made for the materials.

Keywords: Bottom ash; Bentonite; Zeolite; Heavy metal removal; Leachate

1. Introduction

Solid waste landfills include a number of heavy metals and organic pollution. Heavy metals may enter the aquifer and result in potential danger to human and ecological health. Based on the researches, almost all types of landfill liners can leak and generate a danger to the underlying aquifer. The US-EPA has applied some regulations to prevent this danger as early as 1988 [1]. In order to prevent the leakage of contaminants from leachate, clay liners are commonly used in landfills. Conventional liners are designed to minimize leakage of leachate through the liner. However, recent results of environmental monitoring point out that landfill may still have leakage because of construction and operational problems [2]. Adsorption of landfill leachate compounds in clay soils has been studied in recent years and a remarkable amount of data has been found for different soils and compounds. It is argued that the worst contamination is observed in the first years of landfilling so that the contaminants of leachate become less dangerous after some years [3]. However, it is opposed that landfill leachates deliver unacceptable concentrations of contaminants to the environment for periods longer than the life of protective liners [4]. Clay or other liner should be used to prevent toxic contaminants and supply low hydraulic conductivity in engineered landfills. However, heavy metals can still pollute the groundwater by diffusion through the protective barrier. Therefore, there is a crucial need to develop a new type of landfill liner to effectively prevent diffusion of heavy metals. Strongly adsorbing materials added to the liner to remove pollutants is one of the methods [5–7]. However, researches are still continuing to find affordable materials with high treatment efficiency.

Bottom ash from a thermal power plant was used in our study. Disposal of thermal power plant bottom ashes which are produced in considerable quantities is a major problem in terms of environmental health and Turkey's economy [8]. The presence of heavy metals is a serious problem in the environment due to their high toxicity. Bottom ash prevents release of dangerous heavy metals from leachate. Bottom ash was used as an adsorbent to remove Cu and Ni from wastewater and it is found as an appropriate material [9,10].

*Corresponding author.

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The main objective of this study was to use this material as an impermeable layer, instead of storing it as a waste, in solid waste landfill areas to increase the adsorption capacity and decrease the permeability of the bottom liner of the landfill. In the study, two different groups of materials were used to compare the base materials.

Results of the studies on bottom ash have not supplied satisfactory information about the impact of increasing impermeability capacity in stratified systems for the removal of organic pollution and heavy metals. Therefore, this study was aimed to investigate the removal of organics and heavy metals in the layer formed by mixing bottom ash and zeolite with bentonite. The important point here is to increase the impermeability by using bentonite, which is used in the content of geo-synthetic products and provides a high performance, both in terms of swelling of water and hydraulic conductivity, and therefore to obtain more pollution removal and heavy metal retention. For this purpose, the mixture ratios were studied providing the optimum value of impermeability coefficient and heavy metal/organic pollution retention efficiency in the layer.

2. Materials and methods

In the study, bentonite was mixed with bottom ash and zeolite in the ratios of 20%, 30% and 40%. Bentonite was purchased from a local firm. The falling head permeability tests were applied on these three mixtures of bottom ash and zeolite to determine the lowest permeability coefficient. Fining procedure was conducted by hammering in proctors in order to ensure that the bottom ash achieves the desired permeability value (10^{-7} cm/s). Particle size was reduced to the range of 0.1–2 mm. Bentonite ratio in the mixture was increased to provide impermeability. The analysis was carried out according to the soil mechanics test methods as in Turkish Standards (TS 1900/April 1987). Falling head permeameter apparatus was set up as the permeability mechanism. A total of 16 apparatus was used including an individual mold mechanism for each material mixture of which permeability is to be determined. In the falling head permeameter mechanism, mixtures of both zeolite+bentonite and bottom ash+bentonite were first prepared in different ratios in a separate place (after making them finer into appropriate grain size) and compressed by putting in the molds and then permeability values were measured. To evaluate the heavy metals removal capacity, two types of laboratory tests were performed as the bottom ash+bentonite and zeolite+bentonite with synthetic solution similar to the values of the landfill leachate.

In the second part of the study, the removal of physical, chemical and biological pollutants were analyzed in the leachate taken from Istanbul Odayeri landfill site. By comparing the effluent heavy metal concentrations with the limit values specified in the Water Pollution Control Legislation of Turkey, it was investigated whether or not the mixtures could be used as an impermeable layer in landfill sites.

During the study, the measurements of pH, TSS, TKN, NH_3-N, TP, PO_4-P, NO_2-N, NO_3-N, BOD_5 and COD parameters were carried out according to Standard Methods [11]. COD measurements were made according to open reflux method. TSS parameter was made by gravimetric method. TKN and ammonia nitrogen measurements were done by Kjeldahl method and titrimetric distillation. PO_4-P and total-P measurements were made by coloration after acid digestion. The spectrophotometric readings were performed using Pharmacia LKB brand Novaspec II model spectrometer.

Fe, Pb, Cr, Cu, Cd, Ni, Zn, Mg, Mn and Ca were analyzed with atomic adsorption spectrometer (ATI-Unicam 929 AA spectrometer). The leachate was taken from the landfill site and stored at 4°C (see Appendix Table A1 for leachate parameters). According to the peak search graphs obtained by X-ray diffraction method, bentonite was found to be composed of N-20, N-25, N-26, N-27 and N-28, means 98% smectite, 2% quartz and feldspar minerals. Zeolite was composed of Z-1 and Z-2 zeolite group minerals according to the results obtained by X-ray diffraction. (See Appendix Figs. A1 and A2 for X-ray diffraction peak graphs). The pH of the material was 7.3.

Power plant bottom ash used in this study was taken from Afsin-Elbistan power plant (1360 MW) located in the Southern Anatolia. The power plant combusts 18.6 million tons lignite coal per year and generates bottom ash with the quantity of 4.96 million tons per year. The pH value of the material was 11.8 (see Appendix Table A2 for the concentrations of organic and chemical parameters for bentonite, zeolite and bottom ash).

3. Results and discussion

In the first stage of the study, the most appropriate permeability value for each mixture was obtained. The falling head permeameter trials were applied to bottom ash and zeolite mixtures for which separate bentonite additions were made. The lowest permeability coefficient was found to be provided by the mixture with 30% ratio of bentonite material (see Appendix Table A3 for average permeability values of bottom ash, zeolite and bentonite mixture).

Heavy metal retention capacities of the liner materials were determined by filtering in a synthetic solution including heavy metals. This solution was prepared as simulated leachate in the laboratory before filtering the leachate taken from the landfill site. Two separate preliminary experiments were performed for 5 and 10 mg with synthetic solution of heavy metals. However, when 10 g was used, heavy metals were below the reading range and experiments were continued with 5 g to see the reduction ratios clearly. As shown in Table 1, the adsorption values of all other metals except Ca were higher with bentonite and bottom ash.

Chemical affinities of heavy metals in synthetic water were as Pb > Cr > Ca > Cu > Fe > Ni > Zn for zeolite; Fe > Pb=Ni > Cu > Cr > Zn > Ca for bentonite; Fe > Pb=Ni=Zn > Cu > Cr > Ca for the bottom ash. Comparison of the heavy metal adsorption ratios with previous studies are shown in Table 2. As seen in the table, particularly Zn, Pb and Ni were removed with higher efficiency in bottom ash and bentonite materials.

The process of filtering by the leachate taken from Odayeri landfill through the prepared bottom material continued

Table 1
Effluent heavy metal concentrations with the synthetic water (as mg/L)

Heavy metals	Synthetic solution concentration	Effluent concentration		
		Zeolite	Bentonite	Bottom ash
Pb	1	< 0.01	< 0.01	< 0.01
Ni	1	0.62±0.4	< 0.01	< 0.01
Cu	30	11.4±2	0.07±0.004	0.06±0.002
Cr	10	1.1±0.08	0.09±0.005	0.15±0.03
Fe	50	26.3±0.2	0.025±0.002	< 0.01
Zn	1	0.82±0.06	0.05±0.005	< 0.01
Ca	245	43.1±3.5	27.5±3.1	160±11

Table 2
Comparison of layer materials with previous studies (as %)

Parameter	Literature	values	This study		
	Bottom ash*	Bottom ash**	Bottom ash	Zeolite	Bentonite
Zn	82.2	99.99	≥95	≥95	≥95
Mn	94.3				
Fe	96.5		≥95	83-86	97-98
Pb		74.49	≥95	64-66	≥95
Ni		98.9	≥95	55-57	≥95

* [12], ** [13]

Table 3
Effluent parameters and concentrations after filtering the leachate through the bottom material

Parameters	Leachate (Inlet, mg/l)	Effluent concentration, mg/l					
		Set A*			Set B**		
		20% (A1)	30% (A2)	40% (A3)	20% (B1)	30% (B2)	40% (B3)
pH	7.38±0.5	11.87±1.2	9.7±0.87	8.07±0.6	7.37±0.9	7.51±0.84	7.68±0.64
Conductivity	43400±1200	4010±385	1800±155	1770±114	3240±135	5030±442	6240±428
COD	24950±2550	2900±272	2400±187	1110±87	75±6.2	140±11	415±24
TSS	825±218	23±2.1	33±2.9	81±6.4	26±1.4	126±87	85±7
Total P	17.05±1.53	1.95±0.8	1.04±0.9	1.25±0.34	0.7±0.08	1.9±0.6	2.9±0.5
TKN	1036±154	89.6±7.8	67±4.8	8.4±0.53	5.6±0.7	23.8±3.2	36.4±2.7
Fe	85±8	0.14±0.02	0.11±0.02	0.07±0.004	0.89±0.07	0.39±0.01	0.09±0.006
Pb	1.618±0.57	0.07±0.01	0.11±0.02	–	0.1±0.02	0.08±0.006	0.07±0.003
Cr	0.793±024	0.06±0.02	0.01±0.004	–	0.04±0.002	0.07±0.004	0.08±0.004
Cu	0.876±0.31	0.06±0.015	0.03±0.001	–	0.01±0.007	0.02±0.009	<0.01
Cd	0.115±0.06	0.01±0.004	<0.01	–	<0.01	0.02±0.008	0.03±0.005
Ni	1.703±0.52	0.07±0.005	0.04±0.002	–	0.02±0.006	0.04±0.009	0.06±0.005
Zn	1.287±0.47	0.1±0.06	0.19±0.01	–	0.29±0.07	0.25±0.05	0.4±0.07
Mn	9.101±1.3	0.03±0.001	0.07±0.004	–	0.54±0.08	0.6±0.006	1.57±0.3

*Bentonite mixture+ Bottom ash (%),
**Bentonite mixture+ Zeolite (%)

for six months and heavy metals in permeated water were analyzed at the end of six months in two different systems (Set A and Set B in Table 3).

As shown in Table 3, the inlet pH value of garbage leachate is 7.38 while the pH value is lower when the bentonite addition is increased depending on the percent of bentonite mixture due to the high pH value of the bottom ash (pH = 11.8) in the Set A. On the other hand, as the pH value of zeolite (pH = 7.3) is close to the value of leachate, the lower the bentonite (pH = 8.7) addition the lower the pH value is. It is seen that the value of electrical conductivity decreases as the pH value decreases. The electrical potential in the grains is reduced due to the lower pH value [14]. The electrical conductivity in the Set A and B remains below 4000 microsiemens/cm and the salinity value also reduces in this way. In another study with clinoptilolite, the most common zeolite type in the nature, it is found that Pb and Cd removal was very high in low pH, but maximum 35% Cr and Phenol removal was achieved [15].

The COD, TSS, Total P and TKN results obtained for the Set A and Set B show an efficiency of over 80%. When we compare with literature values, there are satisfactory values in both sets. Table 4 shows the highest and lowest efficiencies obtained in 3 separate tasks for each set. Although the results of the biological parameters show that the mixture of Set B has better results than Set A, the Set A provides an incremental removal which should not be underestimated when compared with literature (Table 4).

Results of the experiments for different amount of bottom ash for the removal of metal pollution in leachate are given in Table 4. While Lin and Yang [12] achieved 70–95% success in removal of Fe, Zn and Mn, the success rates were generally between 90–98% ranges in this study. When addressing all available data, it was observed that a remarkable success was achieved, particularly in the removal of Pb, Zn, Cr metals when compared with the prior studies, in terms of heavy metal adsorption. The highest and lowest removal efficiencies of metals for each set are compared with the literature in Table 4.

In recent years, adding materials to the landfill liners, capable of strongly adsorbing pollutants, have been tried by some researchers to increase adsorption capacity of the layers. Lu [7] studied adsorption of Cr(VI) in landfill liners by granular activated carbon (GAC), bentonite activated by acid and natural clay. The adsorption capacity was higher in the order of clay+GAC(3%)>clay+activated bentonite(3%)>natural clay. Also, natural zeolite was used successfully as a liner material with bentonite for the removal of Cu(II) from leachate in landfill [16]. Similarly, addition of shale improved the adsorption capacity of shale-clay mixtures for Zn, Cd, Pb and Cr [17]. It can be seen that adding materials can effectively prevent the transport of some or all of the heavy metals.

Also, it is clear that adding materials can increase the effect of adsorption. Although, Yao [18] could not find a high positive effect by using municipal solid waste incinerator bottom ash; heavy metal removal of Pb, Zn and Ni by using both commercial zeolite and after coal fly ash alkaline hydro-thermal process was analyzed in two different studies and it was reported that a significant increase in terms of the efficiency of COD removal was obtained [19]. Pivato and Raga [1] showed that bentonite added to soil in composite or multi layered liner produced swelling of the particles when hydrated, hence reduced permeability of the soils. In another study, adsorption capacities of Ca-Bentonite, Na-Bentonite and the materials forming natural zeolite were investigated and adsorption values of zeolite to remove $Pb(NO_3)_2$ were found rather low as compared to Ca-Bentonite and Na-Bentonite [20].

A comparison was done to find whether the effluent complies with the standards specified in the regulation on solid wastes (solid waste disposal facilities, leachate effluent limit values). This data also shows that the landfill area does not need an additional treatment unit and therefore it is advantageous with respect to reduce operating costs considerably. In particular, the heavy metal removal ratios are within the first-class water quality standards of the Ministry of Environment Water Quality Classification (see

Table 4
Comparison of removal efficiencies (as %)

	This study		Literature values				
	Set A	Set B	With coal fly ash[a]	With bottom ash[b]	Leachate[c]	Leachate[d]	Artificial zeolite[e]
COD	88–96	97–99	10.3	≤50	–	–	–
TSS	90 –97	85–97	–		–	–	–
Total P	88–94	83–96	100	≤92.9	–	–	–
TKN	91–98	96–99	50.4	≤31.1	–	–	–
Fe	97–98	97–98	–	–	82.5	≤96.5	–
Pb	93–96	93–96	–	–	–	–	>80
Cr	92–98	89–94	–	–	–	–	–
Cu	93–96	97–98	–	–	–	–	–
Cd	>91	73–>91	–	–	–	–	>80
Ni	95–97	96–98	–	–	–	–	–
Zn	85–92	82–94	–	–	60.5	≤ 82.2	>80
Mn	97–98	82–94	–	–	24.3	≤94.3	>80

[a][19]; Diluted original leachate and with zeolitized coal fly ashes
[b][12]; Original leachate and with coal bottom ash
[c][19]; Diluted leachate
[d][12]; Leachate and with artificial zeolite
[e][12]; Leachate and artificial zeolite generated from coal fly ash

Appendix Table A4 for the comparison of the results with the regulation in Turkey).

At the end of the study, a cost evaluation was made for the materials. One of the positive aspects of the product described in this paper is precisely that it is a cheap material derived from a by-product, power plant coal bottom ash. The basic cost component of bottom ash is only shipping, which could be considered competitive with regard to other commercial adsorbents. In this study, the total market cost was found as max. 100 $/ton for zeolite and max. 110 $/ton for bentonite, which could be considered competitive with regard to bottom ash. There will be no cost for the bottom ash but only transportation cost to the waste landfill site.

4. Conclusion

The disposal of municipal solid wastes in sanitary landfill produces leachate that has high concentrations of COD, inorganic pollutants and heavy metals. Physical, chemical and biological treatment processes are widely used for reducing the strength of leachate pollutants. Therefore the application of low cost and easily available bottom ash using landfill liner is an attractive option. In all conducted research studies, no scientific data was achieved especially in terms of the impermeability of the bottom ash and its potential for serving as layer material. From this point in the study, it was achieved that impermeability characteristic of Afsin Elbistan Thermal Power Plant Bottom Ash was improved by appropriate binding material and proper granulometry distribution.

A rather high performance was obtained for effluent parameters concentration of the garbage leachate taken from the landfill site after it was filtered through the two different bottom materials. It was determined that pollutant parameters in the effluent water are much lower than the discharge limit values of the effluent leachate for Solid Waste Recycling and Disposal Facilities' Regulation (Ministry of Environment, Regulation on wastewater discharge standard and ISKI-Istanbul Water and Sewerage Administration, wastewater discharge limit values for in and outside of the basin). It can be clearly seen that using the material obtained from the mixture of bentonite and bottom ash as the bottom material in dumping sites performs well and treats more effectively than the layer formed by zeolite and bentonite in the removal of heavy metals and TSS. In addition, as the bottom ash only has a transportation cost and transform the waste into an economical value, its use has become desirable when compared to zeolite and other materials.

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Appendix

Pollution removal from leachate using bottom ash-bentonite-zeolite liner

Table A1
Composition of Odayeri landfill leachate

Parameter (mg/L)	Range	Parameter (mg/L)	Range
pH	7.2–8	Cu	0.01–0.05
COD	10000–26500	Fe	44.80–58.35
BOD_5	6600–15900	Zn	0.13–0.36
VSS	480–1270	Pb	0.05–0.08
Cond. (ms/cm)	29–33	Ni	0.39–0.78
TKN	2410–2950	Cd	<0.01–0.03
NH_3-N	2070–2730	Alkalinity ($CaCO_3$)	6200–15750
Org-N	30–400	Ca^{2+} hardness	500–700
Total P	17–37	Cl^-	4500–7000
Cr	2.13–3.42	T. Hardness ($CaCO_3$)	2400–4000

Table A2
Chemical composition of bentonite, zeolite and bottom ash (as mg/kg)

Parameter	Bentonite	Zeolite	Bottom Ash
Org. material (%)	12.65	8.84	6.77
Pb	24	44	30
Cr	45.2	4.4	404.8
Zn	58.8	77.2	43.2
Ni	70.8	1.6	108.4
Cu	24.8	4.4	37.6
Fe	14000	2560	18500
Mn	236	130	176

Table A3
Permeability in bottom ash, zeolite and bentonite mixture

Used layer materials	Permeability values in Bentonite mixture ratios (%)*				
	% 0	% 10	% 20	% 30	% 40
Bottom ash + %bentonite	2.1×10^{-3}	5.5×10^{-5}	6.2×10^{-7}	2.1×10^{-8}	8.1×10^{-8}
Zeolite + %bentonite	3.5×10^{-3}	7.1×10^{-4}	2.1×10^{-6}	1.1×10^{-7}	3.2×10^{-7}

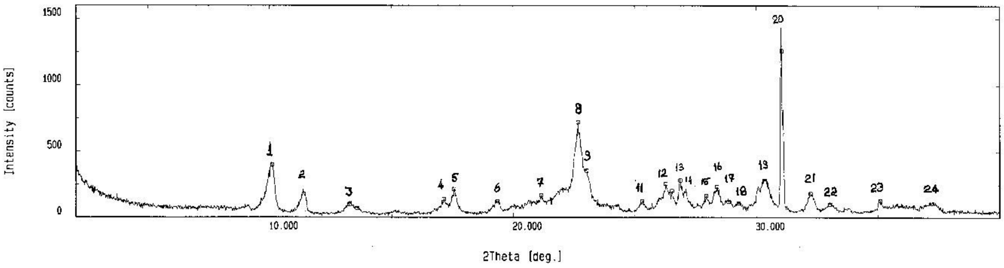
* Grain diameters: 0.1–2 mm

Table A4
Comparison of the results with the regulation in Turkey

Parameter (mg/L)	Bottom ash+%30 Bentonite	Zeolite +%30 Bentonite	Composite sampling	
			2 h	24 h
COD	2400	140	160	100
TSS	33	126	200	100
PO_{4-P}	1.04	1.9	2	1
Total Cr	0.01	0.07	3	1
Pb	0.11	0.08	2	1
Cd	0.01	0.02	0.1	–
Fe	0.11	0.39	10	–
Cu	0.03	0.02	3	–
Zn	0.19	0.25	5	–
pH	9.7	7.51	6–9	6–9

Peak search of zeolit

Sample	: z1	[Smoothing]	
File	: 1.0260	[B.G. elimination]	
Comment	: 1	[Ka2 elimination]	
Date	: 28-Jun-01 01:17	[Peak search]	Typical width : 0.30 Minimum height : 30.000
Operator	: RINT		
Memo	: z1		

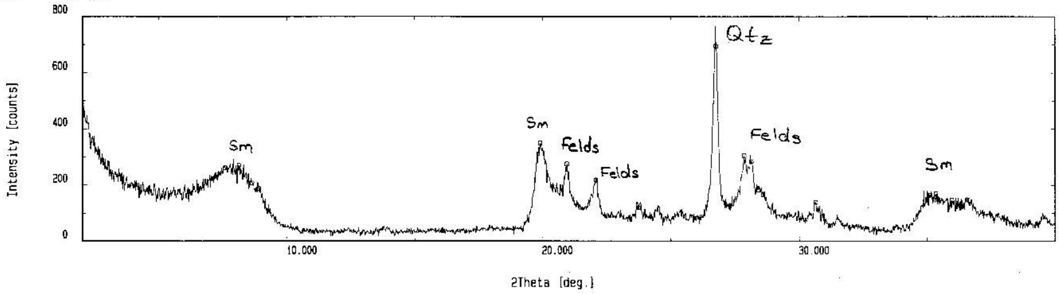


Peak No.	2Theta	FWHM	d-value	Intensity	I/Io
1	10.100	0.329	8.7507 zeo	399	32
2	11.400	0.282	7.7556 zeo	176	14
3	13.260	0.329	6.6716 zeo	102	10
4	13.580	0.188	6.5151 zeo	70	6
5	17.140	0.188	5.1691 zeo	140	12
6	17.560	0.235	5.0464 mica	216	18
7	19.340	0.235	4.5857 Pyr	126	10
8	21.140	0.188	4.1992 K-Felds	170	14
9	22.660	0.353	3.9208 zeo	722	58
10	23.020	0.212	3.8603 zeo	356	30
11	25.300	0.212	3.5173 zeo	126	10
12	26.280	0.212	3.3884 zeo	257	22
13	26.540	0.165	3.3558 Pyr	204	18
14	26.880	0.188	3.3141 K-Felds	282	24
15	27.940	0.235	3.1907 K-Felds	169	14
16	28.380	0.306	3.1422 Zeo	234	20
17	28.860	0.306	3.0911 Pyr	120	10
18	29.280	0.282	3.0477 Pyr	110	10
19	30.340	0.329	2.9436 Zeo	280	24
20	31.040	0.165	2.8788 Zeo	1260	100
21	32.220	0.329	2.7760	184	16
22	33.020	0.306	2.7105	100	8
23	35.080	0.212	2.5559	132	12
24	37.200	0.353	2.4150 Zeo	112	10

Fig. A1. Zeolite X-Ray diffraction peak graph.

Peak search of bentonit

Sample	: n-27	[Smoothing]	
File	: 1.0257	[B.G. elimination]	
Comment	: 1	[Ka2 elimination]	
Date	: 27-Jun-01 23:55	[Peak search]	Typical width : 0.30 Minimum height : 30.000
Operator	: RINT		
Memo	: n27		



Peak No.	2Theta	FWHM	d-value	Intensity	I/Io
1	7.580	0.165	11.6533	263	38
2	8.120	0.165	10.8795	272	40
3	8.620	0.165	10.2495	209	30
4	8.880	0.165	9.9500	189	28
5	19.900	0.424	4.4579	351	52
6	20.940	0.212	4.2388	277	40
7	22.060	0.212	4.0261	219	32
8	23.740	0.259	3.7448	120	18
9	26.740	0.235	3.3311	696	100
10	27.840	0.188	3.2019	307	46
11	28.120	0.188	3.1707	285	42
12	28.400	0.165	3.1401	184	28
13	28.580	0.235	3.1207	168	26
14	30.640	0.165	2.9154	140	20
15	35.100	0.212	2.5545	167	24
16	35.340	0.165	2.5377	172	26
17	35.960	0.188	2.4954	148	22
18	36.560	0.165	2.4558	150	22

Fig. A2. Bentonite X-Ray diffraction peak graph.