

## Behavior of nanobentonites in adsorption of cesium and strontium

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**Abstract.** Bentonites contain a large amount of montmorillonite and this nano-structured and nano-porous member of the smectite group determines the adsorption properties of bentonites. In this study, four types of Iranian commercial bentonite clays have been examined to determine the adsorption capacity of cesium and strontium. The physicochemical and mineralogical properties of the four types have been experimentally analyzed with the focus on the moisture content, pH, dry (granulometric) and wet screen analyses; swell, sediment and Methylene Blue (M.B.) indices; cation exchange capacity (C.E.C) measurements. The pH and C.E.C values of B3 were 9.48 and 71.9 mEq/100g, respectively. The high values of C.E.C; swell, sediment and M.B. indices along with the high amount of granulometric values (particles finer than 36  $\mu\text{m}$ ) showed that the B3 had a high amount of montmorillonite. X-Ray Fluorescence (XRF) analysis showed that the major of competing cations such as  $\text{Ca}^{2+}$  and  $\text{K}^{+}$  existing in B2 affects the Sr adsorption. The experimental analysis also showed that B2 had almost high pH and C.E.C values as well. The results of the separation process of Cs and Sr indicate that the B3 and B2 have the highest adsorption capacity of 59.75 and 45.5 for Cs and Sr adsorption, respectively.

**Keywords:** adsorption; metal removal; montmorillonite; nano-structured bentonite

### 1. Introduction

Bentonite as a natural clay with montmorillonite as its dominant constituent part is the result of erosion and degradation of volcanic ashes which mainly take place in swampy environments somewhere away from the volcano where the bentonite thickness reduces. It is an aluminosilicate with a three-layer silicate structure (Ghazizahedi and Hayati-Ashtiani 2018). Bentonites in general are of two major types namely Sodium type with high water swell ability and swell up and Calcium type with high water swell ability and low inflation, swell down and high cation exchange capacity. Swell ability, high cation exchange capacity, and low inflation are some features which make bentonite a good adsorbent for the removal of heavy metals from wastewaters. Also, bentonite can be used by itself as an adsorbent, or in combination with other crushed rock (Wan *et al.* 2020).

Heavy metals as metallic chemical elements are poisonous and as some toxic chemical pollutants are, even at lower concentrations, potentially very harmful to the environment and human health. These metals cannot be easily removed from the human body and can be accumulated in the tissues and resultantly can lead to cancer-related deaths (Marz 1997, Baylan and Meriçboyu 2016). Most studies show the fact that these harmful pollutants are substantially generated from the industrial activities. Some of these metals such as cesium and strontium are produced from nuclear fission, and medical,

agricultural, and environmental activities. So far, some methods such as adsorption, electrodialysis, reverse osmosis (RO), ultrafiltration, solvent extraction, chemical precipitation, ion exchange, and nano-technics have been introduced for the removal of heavy metals from different wastes. Among these methods, adsorption is more important and practical one since this method has high efficiency, produces no toxic chemicals, and is not a costly and time-consuming process (Saleh *et al.* 2019). In this method, bentonite functions as a useful adsorbent for cesium and strontium removal from wastewaters because it has a high stability against radioactive rays, a high specific surface area, a high cation exchange capacity, and a high swell ability (Gunatilake 2015, Abou-Lilah *et al.* 2020).

The properties of bentonites as an excellent adsorbent for removing metals such as cesium and strontium have been experimentally examined in some previous studies. For instance, the study of pH effect showed that the adsorption of cesium increased from 55 to 87% while the pH increased from 2 to 7 and similarly, the adsorption of strontium increased from 46 to 95% which might be due to an increase in the number of available sites (Seliman *et al.* 2014, Khan 2003). The dry screen analysis showed the portion having finer particle size had a higher amount of montmorillonite; consequently, it increased the adsorption capacity of cesium and strontium (Hayati-Ashtiani 2011, Choi *et al.* 2001). The Methylene blue method for determining the specific surface area is better than BET method since both the internal and external surface areas using M.B. method can be calculated through this method. The high specific surface area indicated the high amount of the adsorbent montmorillonite (Hayati-Ashtiani *et al.* 2011, Kuleshova *et al.* 2014). Studies showed the adsorption capacity of Cs and Sr increased with an increase in C.E.C

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values and the results showed the higher amount of montmorillonite in adsorbent resulted in the high swelling index (Ostovaritalab and Hayati-Ashtiani 2019, Hayati-Ashtiani 2012). Other experiments including wet screen, sediment index, and moisture content were used for determining the impurities of bentonites (Hayati-Ashtiani *et al.* 2011).

This research aims at studying the properties of bentonites for adsorption of cesium and strontium using experimental analyses. These analyses include the effect of pH, moisture content, dry and wet screen analyses; swelling and sediment indices; C.E.C and M.B. surface area measurements. The first novel insight provided was the above-mentioned characterization experiments which yielded some significant results which showed the adsorption characteristics of the best type of bentonite to be used for the removal of Cs(I) and Sr(II). Further, since the mechanism of Cs(I) adsorption is different from that of Sr(II), these experiments have been applied with different new insights.

## 2. Materials and methods

Four types of Iranian commercial bentonite clays, namely, B1 to B4, in their natural forms were used in this study. First, their physio-chemical and mineralogical properties were exactly determined. Subsequently, through appropriate experimental analyses, the amount of adsorption of cesium and strontium onto bentonite clays was determined. Cesium(I) nitrate ( $\text{CsNO}_3$ , purity  $\geq 99\%$ ) and Strontium(II) nitrate ( $\text{Sr(NO}_3)_2$ , purity  $\geq 99\%$ ), Methylene Blue ( $\text{C}_{16}\text{H}_{18}\text{ClN}_3\text{S}\cdot x\text{H}_2\text{O}$  ( $x=2-3$ ),  $50\% \leq$  purity  $\leq 100\%$ ), and sodium phosphate dodecahydrate ( $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$ , purity  $\geq 98\%$ ) were purchased all from Merck Chemicals.

### 2.1 Sampling, drying and ball-milling treatment

At the site, the air-dried samples were ground with crushers. For better grinding, the samples had to be further dried in an oven (Fan Azma Gostar V.2.1, Iran) and then further ground with a laboratory jar and ball mill to powder of less than  $75\ \mu\text{m}$ . The name, origin, and supplier of the bentonites are given in Table 1.

### 2.2 Moisture content

The general purpose of this test was to determine the amount of free moisture in the samples. 100 grams ( $W_1$ ) of each sample having been passed through mesh No. 200 was weighed with 0.0001 g precision. Four samples were weighed and put into the oven at  $110^\circ\text{C}$  and were weighed at the specified intervals (each half an hour) until they reached a constant weight. When two consecutive points with constant mass were reached, the test was completed. Samples were taken out of the oven and put into the desiccator to be cooled and then they were weighed again ( $W_2$ ). Eq. (1) was used to calculate the moisture content of the samples (ASTM D 2974-00, 2000).

Table 1 The names, origins, and supplier of the bentonite samples

Sample Name	Location of Mine	Supplier
B1	Chah-e Kashmir mine, Birjand, southern Khorasan Province, Iran	Iran Chemiral Co.
B2	GolKan mine, Gonabad, southern Khorasan Province, Iran	Same as for B1
B3	Chah-e Golestan mine, Sarayan, southern Khorasan Province, Iran	Same as for B1
B4	BotehGaz mine, Asaad Abad, southern Khorasan Province, Iran	Same as for B1

$$\text{Moisture content (\% wt)} = \frac{W_1 - W_2}{W_1} \times 100 \quad (1)$$

In Eq. (1),  $W_1$  is the wet bentonite weight (g) and  $W_2$  is the dry bentonite weight (g).

### 2.3 pH effects

The main purpose of this test was to determine the amount of alkaline of the natural bentonites. Two grams of the dried bentonite powder were weighed with 0.0001 g precision. The powder was gradually added into the beaker containing 100 mL of distilled water. The suspension was then sufficiently mixed for at least 5 minutes so that a uniform suspension could be formed. Later on, the pH of the solution was measured with the pH meter. When the pH meter indicated a fixed number, it was kept in solution. Also, the electrode of pH meter was kept in the partially settled suspension so that the effect of collision of particles in the suspension could be diminished (ASTM D 4972-01, 2001).

### 2.4 Dry screen (granulometric) analyses

This test was conducted so that the particle size distribution of bentonites could be determined. 100 grams of dried bentonite powder was weighed with 0.0001 g precision. It was sieved to some different mesh number sieves in ascending order. The sieves were shaken by the vibrator until no further material could pass through the sieves. Then, the remaining amount on each sieve was carefully weighed with 0.0001 g precision (API 1994, ASTM D 4381-01).

### 2.5 Wet screen analysis

This test was carried out so that the amount of water-insoluble material like sands not passing the sieve mesh No. 200 could be determined. Then, 14 grams of each bentonite sample ( $W_3$ ) was weighed with 0.0001 precision and slowly poured over the 350 ml of water in the beaker which was stirring with a magnet stirrer for 15 minutes. Subsequently, 0.2 g of sodium phosphate dodecahydrate was added to the suspension and the suspension was stirred for another 5 minutes. The suspension was poured into the sieve mesh No. 200 ( $74\ \mu\text{m}$ ) previously wetted with water (ASTM E 11-70) and the residue on the sieve was washed

with a mild stream of tap water so that no further materials could pass through. The sieve and residue were then put into the oven for two hours at 110 °C. Next, the residue on the sieve was transferred to the watch glass ( $W_2$ ) and weighed to 0.1 precision and the watch glass along with the remaining on the sieve was weighed with 0.01 g precision ( $W_1$ ). Finally, the remaining amount, with the precision of 0.1 percent, was obtained through the Eq. (2) (ASTMD 4381-01 2001; ASTM C 117-95 1995):

$$Wt\% \text{ of remaining amount (Amount of sand)} = \frac{W_1 - W_2}{W_3} \times 100 \quad (2)$$

In Eq. (2),  $W_1$ ,  $W_2$  and  $W_3$  are watch glass plus residue (g), watch glass (g) and bentonite sample (14 g) weights, respectively.

## 2.6 Swell index

Through this test carried out based on ASTM D 5890-02, the amount of adsorbed water causing the swelling of bentonite clays could be determined. The bentonite samples were powdered so that they could fully (100%) pass through sieve mesh No. 100 (149  $\mu\text{m}$ ) and at least 65% through sieve mesh No. 200 (74  $\mu\text{m}$ ). The swell index was attained by gradually addition of a 2-g sample in about 0.1 g increments to 90 mL of water in a 100 mL graduated vessel. The hydrated bentonite columns for trapped air release or water separation were examined after 2 hours from the last addition. The swell index values corresponding to the volume of hydrated samples were registered in mm after at least a 16 -hour hydration from the last increment addition.

## 2.7 Sediment index

Sediment volume as an influential factor had to be determined through sediment index test. In this stage, one gram of each sample was weighed and poured into a beaker containing 90 mL of water. The suspension was vigorously stirred for five minutes until a homogeneous suspension was obtained. 10 mL of a normal solution of ammonium chloride ( $\text{NH}_4\text{Cl}$ ) was then added to the suspension and the mixture was thoroughly stirred again. Finally, the volume of sediment was measured after 72 hours and the sediment index was obtained from the Eq. (3) (Hayati-Ashtiani *et al.* 2011):

$$SI = \frac{V}{100 - M} \times 100 \quad (3)$$

## 2.8 Surface area measurements

The specific surface area of bentonite was also calculated through the methylene blue-spot test with methylene blue chloride powder. In this step, one gram of dried powder of M.B. was mixed with 200 mL of distilled water. Then, 10 g of dry bentonite sample was mixed with 30 mL of distilled water. Each time 0.5 mL of M.B. solution was added to the bentonite suspension and was mixed with all the bentonite suspension together thoroughly

for 1 min. Then, a small drop of the suspension was put on a filter paper and the specific surface area was determined from the amount of M.B. required to reach the endpoint of the test. The endpoint was reached when the unabsorbed M.B. formed a permanent light blue halo around the soil aggregate spot. To add 0.5 mL of methylene blue solution was low in each step with high C.E.C bentonites. Therefore, 20 mL of methylene blue solution was added in each step and the suspension was mixed for 40 minutes. The relation between surface area and methylene blue consumption was calculated through the Eq. (4) (Santamarina *et al.* 2002, Yukselen and Kaya 2006):

$$SSA = \frac{1}{319.87} \frac{1}{200} (0.5N) A_v A_{MB} \frac{1}{10} \quad (4)$$

Where SSA is the specific surface area, N is the number of the added M.B. to the suspension,  $A_v$  is Avogadro's number ( $6.02 \times 10^{23}$ ), and  $A_{MB}$  is the area covered by a molecule of M.B. (it is usually assumed as 130  $\text{\AA}^2$ ).

## 2.9 Cation exchange capacity (C.E.C) measurements

Determining the cation exchange capacity by M.B. is really essential in this study and the like. The procedure followed in this regard was exactly like that of M.B. test. After determining the total volume of consumed solution, cation exchange capacity was obtained from the Eq. (5) (Hayati-Ashtiani *et al.* 2011):

$$\begin{aligned} \text{CEC} = \text{MB Added (mL)} \times \frac{\text{MB Dry Wt(g)}}{319.87} \\ \times \frac{100}{\text{Vol. of MBSolution}} \times \frac{100 \text{ g}}{\text{Clay Dry Wt(g)}} \text{ (mEq/100g)} \end{aligned} \quad (5)$$

Since the amount of clay dry and M.B. dry is 1 and 10 g, respectively, the Eq. (5) could be simplified as Eq. (6) (Hayati-Ashtiani *et al.* 2011):

$$\text{CEC} = \frac{\text{MB added(mL)} \text{ Wt(g)}}{\text{Vol. of MB Solution(mL)}} \times \frac{1000}{319.87} \text{ (mEq/100g)} \quad (6)$$

## 2.10 Separation process of Cs and Sr

Two solutions, one of them with Cs(I) and the other with Sr(II) at 1000 ppm concentration, were prepared so that to study Cs(I) and Sr(II) adsorption could be examined. All glassware and plastic bottles were carefully washed with distilled water and then rinsed with acetone solution. Then, 0.3 g of adsorbent was added to 25 mL of Cs and Sr solutions in plastic bottles and then the suspension was mixed using a shaker (INFORS AGCH-4103 BOTTMINGEN). The shaker time and revolution were fixed to 24 h and 150 rpm, respectively. The effects of equilibrium times on the removal of the Cesium and Strontium were eliminated when the shaking times were fixed to 24 h. After 24 h, the suspension was filtered with a vacuum pump to separate the adsorbents from the solution. Next, 1 to 2 mL of the solution containing Sr(II) or Cs(I) was sent to ICP laboratory (Varian Corp., Liberty 150AxTurbo, Australia) for measuring metal adsorption. The samples containing Cs(I) were analyzed by means of an atomic absorption

spectrophotometer (Spectra AA200 model, Varian Corp., Victoria, Australia) with air-acetylene flame. To calibrate the instruments, 2, 5, 10 and 20 ppm solutions were used by diluting the stock solution having 1000 ppm concentration. Calibrations were made fresh each time before any analysis.

Distribution ratio, Adsorption percentage and Adsorption capacity were calculated as Eqs. (7)-(9), respectively. (Ostovaritalab and Hayati-Ashtiani 2019):

- Distribution ratio

$$K_d = \frac{C_0 - C_{eq}}{C_{eq}} \cdot \frac{V}{m} \quad (7)$$

- Adsorption percentage

$$R = \frac{100 \cdot K_d}{K_d + \frac{V}{m}} [\%] = (1 - C/C_0) \times 100 \quad (8)$$

- Adsorption capacity

$$\Gamma = K_d \cdot C = (C_0 - C) \cdot V/m \quad (9)$$

$K_d$  is distribution coefficient ( $\text{mLg}^{-1}$ ),  $C_0$  is the initial concentration ( $\text{gL}^{-1}$ ),  $C$  is the equilibrium concentration ( $\text{gL}^{-1}$ ),  $V$  is the aqueous phase volume (mL),  $m$  is the adsorbent mass (g),  $R$  is the adsorption percentage, and  $\Gamma$  is adsorption capacity.

### 3. Results

#### 3.1 Moisture content

Enough information about of moisture content would be very useful to determine the desired time for each specific sample to become anhydrous solid under a drying condition. Table 2 indicates that B3 and B1 have the highest and lowest moisture content, respectively.

#### 3.2 pH effects

The results of the pH effects are reported in Table 2. The pH has almost a direct relation to cation exchange capacity; i.e., the C.E.C increases by increasing the pH. All the samples are in the alkaline pH range and the alkalinity degree of sample B3 is more than those of the other samples.

#### 3.3 Dry screen analysis

This analysis shows the portion of fine particles like montmorillonite in samples. Table 3 shows that B1 and B3 have the maximum number of fine particles and B4 and B2 have the maximum number of coarse particles among the other samples.

#### 3.4 Wet screen analysis

This analysis shows the portion of insoluble particles such as silica and sand in samples. The results of the analysis are reported in Table 4. Sample B2 and B3 have the highest and lowest amounts of insoluble particles among the samples, respectively.

Table 2 The moisture content and pH effects of samples

Sample name	pH	Moisture content (Wt%)
B1	8.81	12.639
B2	8.67	6.612
B3	9.48	6.311
B4	8.83	10.008

Table 3 Dry screen analysis of raw bentonites

Mesh No.	-400	-270+400	-200+270	-140+200	+140
B1	63.751	8.604	19.967	5.086	1.687
B2	35.376	9.014	31.178	13.174	10.169
B3	40.207	10.728	34.189	12.773	2.852
B4	31.045	6.864	6.864	23.813	15.236

Table 4 Wet screen analysis of raw bentonites

Sample name	Wt% of residue (sand)
B1	2.28
B2	7.47
B3	1.057
B4	4.25

Table 5 The swell, sediment and Methylene Blue (M.B.) Indices and Cation exchange capacity (C.E.C) of samples

Sample name	C.E.C. (mEq/100g)	Indices		
		Swell (mm)	Sediment (mL/g)	M.B. ( $\text{m}^2/\text{g}$ )
B1	56.27	5	18.315	440.39
B2	50.02	8	24.628	391.46
B3	71.9	28	80.052	562.72
B4	78.16	25	81.118	611.65

#### 3.5 Swell index

The swell index measurement is one of the best methods to determine the rate of water adsorption during the metal removal process. The results of the swell index in mm are shown in Table 5. In this analysis, B3 and B1 have the highest and lowest swell index, respectively.

#### 3.6 Sediment index

The sediment index provides us with a convenient means of sedimentation analysis of samples. The results of the sediment index for bentonite samples are reported in Table 5. Sample B4 and B1 have the highest and lowest sediment index values, respectively. Sample B3 sediment index has a very close value to that of B4.

#### 3.7 Surface area measurements

M.B. analysis is one of the best and most accurate methods for determining the specific surface areas of bentonites since this method can measure the internal and external surface areas while the other methods like BET can

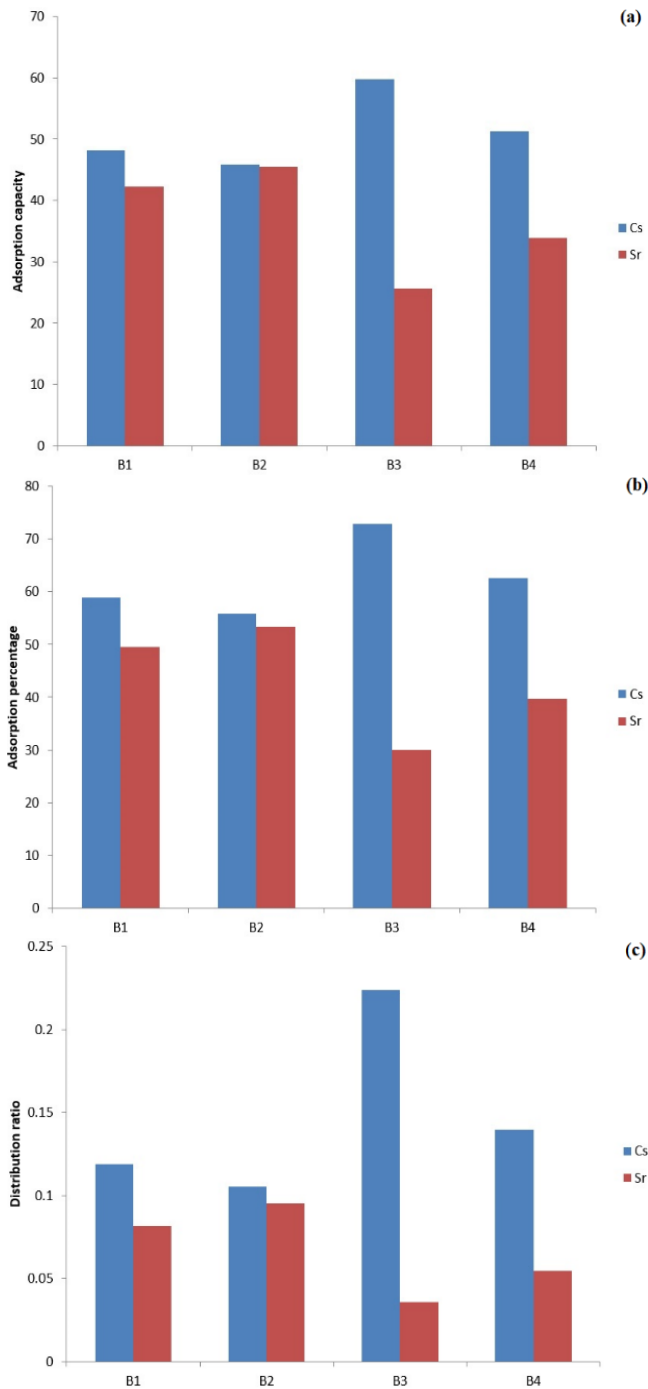


Fig. 1 (a) Adsorption capacity, (b) adsorption percentage and, (c) distribution ratio of samples

only measure the external surface areas of bentonites. The results of the surface area are reported in Table 5. According to this analysis, B4 and B2 have the highest and lowest specific surface areas, respectively.

### 3.8 Cation exchange capacity (C.E.C) measurements

Cation exchange capacity (C.E.C) measurement is one of the methods which help find out the rate of Cs and Sr adsorption of samples. Table 5 shows the highest and lowest values of C.E.C related to B4 and B2 samples, respectively.

### 3.9 Separation process of Cs and Sr

Fig. 1 shows the results of the adsorption capacity, adsorption percentage, and distribution ratio of Cs and Sr adsorption are higher in B3 and B2 than those of the other samples. The Cs and Sr distribution ratio and adsorption percentage have greater values for B3 and B2 samples. Fig. 1 shows the decrease in adsorption capacity ( $K_d$ ), adsorption percentage ( $R$ ), and distribution ratio ( $\Gamma$ ) of Cs and the order is as follows:  $B3 > B4 > B1 > B2$ ; a reverse relation is observed for  $K_d$ ,  $R$  and  $\Gamma$  of Sr adsorption:  $B2 > B1 > B4 > B3$ .

## 4. Discussions

Sample B3 is an excellent adsorbent for heavy metal removal. The high pH value of sample B3 indicates a good cation exchange capacity for this sample. Other studies have shown that most of the bentonite samples were in the alkalinity range of pH (between 7.4–10.4) with different alkalinity degrees (Hayati-Ashtiani *et al.* 2011). The high pH value of sample B3 indicates that more exchangeable cations such as  $\text{Na}^+$ ,  $\text{Ca}^{2+}$  and  $\text{K}^+$  are released into solution during any wet processes and exchangeable cations are the reason for high C.E.C values of bentonites (Table 2 and 5). Therefore, B3 has a good potentiality as the adsorbent for adsorption of Cs and Sr but the separation process test of Cs and Sr showed that this sample has the lowest and highest adsorption capacity for Sr and Cs, respectively. This result demonstrates that the cation exchange capacity mechanism or chemical adsorption is not the only mechanism for adsorption of Sr and Cs; therefore, some other mechanisms including the surface or physical adsorption and the effect of competitive cations such as  $\text{Na}^+$ ,  $\text{Ca}^{2+}$  and  $\text{K}^+$  and etc. must be considered.

Low pH values cause the adsorption capacity of Cs and Sr to decrease. The bentonite structure may be partially collapsed due to the cation extraction into the acidic solution and the available sites for adsorption of metals such as  $\text{Cs}^+$  and  $\text{Sr}^{2+}$  are reduced at  $\text{pH} < 3.5$ . In this study, all samples are in the alkaline range of pH but B2 has a lower pH than that of the other samples; thus, low pH may decrease the adsorption capacity of samples. Another reason for the decrease of the adsorption capacity is competing for  $\text{H}^+$  with  $\text{Cs}^+$  and  $\text{Sr}^{2+}$  since the amount of  $\text{H}^+$  in solution is high at low pH. The amount of Cs adsorption shows the  $\text{H}^+$  has less adsorption competition with Cs in comparison with Sr.

The moisture content is a parameter that has to be considered prior to any experiments because it is a determining factor in drying, grinding or any other preparation processes of the bentonite adsorbents. Table 2 shows that the moisture content for B3 is less than that of the other samples. This result indicates that B3 requires less drying which can be an advantage for sample B3 through decreasing the energy cost and the preparation processes.

Cation exchange capacity has almost a direct relationship with pH and C.E.C. It is noteworthy that the recorded C.E.C. of bentonites is in the range of 28–106

mEq/100 g or higher. The results are in agreement with those mentioned in the literature (Hayati-Ashtiani *et al.* 2011). The high C.E.C. results in more easily releasing of competitive cations into the solution and this will increase the pH of the suspension. Our bentonite samples commonly have high C.E.C. and adsorption capacity (Table 5) since they have an acceptable interlayer exchangeable cation capacity because of its montmorillonite. Therefore, the high amount of montmorillonite will increase the adsorption capacity of bentonite. It is concluded from Table 5 that B4 has the highest amount of C.E.C. with 78.16 mEq/100g, but its pH value is lower than that of B3. This may be due to the presence of impurities in B4. These impurities in addition to lowering the pH value in comparison with B3 may reduce the adsorption capacity of B4. The reduction in adsorption capacity of cesium and strontium of B4 showed in Fig. 1. B3 has a high C.E.C. and good adsorption capacity for cesium. Cesium adsorption on B3 mostly takes place in chemical adsorption since the chemical adsorption has a direct relationship with the C.E.C.

The amount of powder passing through the sieve mesh No.400 (36  $\mu\text{m}$ ) has a direct relationship with the amount of montmorillonite since montmorillonite is basically composed of very fine powder. The results in Table 3 demonstrate that sample B1 has the highest amount of fine powder passing through mesh No. 400. The swell and sediment indices of B1 are low. It is concluded that the reason why the amount of montmorillonite and adsorption capacity for B1 is low is that the existing impurities outweigh the amount of montmorillonite. The amount of the fine powder passing through mesh No. 400 for B4 is the lowest amount among the others and it shows that the impurity constituents such as silica, calcite, etc. are the highest in B4. This claim is confirmed by the results of wet screen analysis. Fig. 1 shows that the large number of the fine particles and small number of insoluble particles will increase the adsorption capacity in the samples. The results of adsorption of cesium and strontium show the adsorption capacity of B1 and B4 is relatively low.

B2 and B4 have the largest number of insoluble particles in water at wet screen analysis. This latest conclusion is simply confirmed with dry screen analysis since B2 and B4 have the highest number of coarse particles. Although, B4 has the largest number of insoluble and coarse particles, the results of other analyses such as C.E.C., swell and sediment indices, and M.B. test approve that B4 has a high amount of montmorillonite in addition to other impurities like insoluble constituents. Based on the results of dry and wet screen analyses, B3 has a high amount of montmorillonite and this result is confirmed with high adsorption capacity of Cs and Sr. Fig. 1 shows that the B3 has the highest Cs adsorption capacity.

The swell index of bentonites has almost a direct relation to C.E.C. Table 5 shows that B3 and B4 have some high swell index values and it means that they have a high amount of montmorillonite. As a consequence, they have the highest amount of montmorillonite. This conclusion is supported by dry and wet screen analyses. Therefore, the Cs adsorption analysis shows that the adsorption capacity of Cs in B3 and B4 is more than that of the other samples.

Montmorillonite has a porous structure and high specific surface area which can be determined with the M.B. test method. The results of M.B. specific surface area measurements are in agreement with the results of the other tests including pH and C.E.C. values and swell and sediment indices, and dry and wet screen analyses. Santamarina *et al.* (2002) showed in M.B.-Spot Test Method (European Standard) that the specific surface areas of montmorillonites (bentonite) were from 364.8-912.0  $\text{m}^2/\text{g}$ . Table 5 shows that B3 (562.72  $\text{m}^2/\text{g}$ ) and B4 (611.65  $\text{m}^2/\text{g}$ ) have the largest specific surface area than those of B1 (440.39  $\text{m}^2/\text{g}$ ) and B2 (391.46  $\text{m}^2/\text{g}$ ). Also, the Cs and Sr adsorption studies show that B3 and B4 have higher adsorption capacity for Cs than those of other samples.

Chemical and physical adsorption has a direct relationship with the cation exchange capacity and specific surface area. Since cesium adsorption is high in sample B3 with a high C.E.C. value, the mechanism of cesium adsorption is cation exchange which is chemical adsorption. The surface area and cation exchange capacity of B2 and B1 show that the competitive cations such as  $\text{Na}^+$ ,  $\text{K}^+$  and  $\text{Ca}^{2+}$  affect the adsorption capacity of Sr.

The competitive cations considerably influence the Cs and Sr separation processes. Adsorption of Sr shows a decrease when univalent cations such as  $\text{K}^+$  and  $\text{Na}^+$  or divalent competitive cations such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  exist in the solution. The presence of these cations may have influenced the adsorption capacity of B2. B2 has almost high pH and C.E.C. (Tables 2 and 5) as well as the XRF analysis shows there is a majority of competitive cations such as  $\text{Ca}^{2+}$  and  $\text{K}^+$  in B2 which easily releases its cations into solution. Competitive cations found between the adsorbent layers have a low concentration in solution since these cations are dissolved in water and few of them will be released to the solution and they will cause a slight increase of the pH of the solution.

Researchers have shown that the distribution coefficient of strontium is maximum in the presence of  $\text{K}^+$  (Galambos *et al.* 2009). Accordingly, the high adsorption capacity of B2 for strontium would be caused by the presence of  $\text{K}^+$  since the XRF analysis shows that the amount of  $\text{K}^+$  in B2 is 0.57 wt% which is the highest amount of  $\text{K}^+$  in all samples.

## 5. Conclusions

Clays are among the cost-effective materials used for removing heavy metals such as Cs and Sr. Bentonite clays play key roles in adsorption processes confirmed by experimental analyses and separation processes of Cs and Sr. Granulometric analysis showed that the B1 had the highest number of fine particles passing through mesh No. 400 (36  $\mu\text{m}$ ) but the presence of impurities decreased the adsorption capacity of sample B1. Wet screen analysis showed that B2 and B4 had some insoluble particles in the water which was confirmed by dry screen analysis. The results of swell and sediment indices, C.E.C. measurements, and M.B test pointed to the conclusion that the B3 and B4 had the highest amount of montmorillonite. The Cs and Sr

had different adsorption mechanisms including chemical, physical, or physicochemical mechanisms. The coexistence of some cations like  $K^+$  changed the adsorption capacity of bentonites. The coefficient of distribution of Cs(I) and Sr (II) depended on the amount of  $K^+$ . Sample B2 had the highest capacity for the adsorption of Cs(I) since it had the lowest amount of  $K^+$  and Sample B3 had the lowest capacity for the adsorption of Sr(II) since it had the highest amount of  $K^+$ .

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