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Manufacturing Method of Cathode Electrode for FEM-EK Process to Adsorb Cesium (Cs) Ion

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ABSTRACT

Soil pollution is one of the main problems after the atomic power plant explosion in Japan. Electrokinetic method can be an important soil remediation method. The method has been proved efficient in laboratory scale, but still requires some solutions to apply it in practical field. However, a new method has been developed, which uses a flat electrode on the surface of the polluted soil as cathode and the anode electrodes are put inside the soil, and then the electrokinetic method is applied in between the electrodes. We have named this process as FEM-EK (Flat Electrode Method-Electrokinetic) process. This new process needs electrodes which can adsorb radioactive materials. We have tried to induce some materials and methods to prepare this new type of cathode electrodes. Natural zeolite, Kuntan (burnt rice husk) and Abaca in different amounts were used to prepare the electrodes. We evaluated the electrodes by measuring the conductivity, flexibility and the adsorption ability of Cs. Our experiments have shown that a combination of zeolite, Kuntan and Abaca may fulfil the needs of the electrode for the FEM-EK process. These electrodes and FEM-EK process can pave the way for purification of radioactive polluted soil in the near future.

INTRODUCTION

After the devastating earthquake in 2011, the explosion of the atomic power plant at Fukushima spread massive radioactive materials all over the Japan, especially near the Fukushima region. The radioactive materials accumulated from the air to the ground surface and stayed there according to their half life period. The half life of radioactive materials depends on the related radioactive materials. From the Fukushima power plant, the main radioactive materials that are responsible for the contamination of the environment were iodine (^{131}I) and cesium (^{137}Cs). The half life of these materials are 8.04 days and 30.07 years respectively. So it can easily be understood that the radioactive material such as Cs will be present in its radioactive form for several years on the ground surface of the soil. And it is obviously a danger to health and life of the living beings.

To remove the radioactive materials from the contaminated soil, in most cases the drilling process was being used in Fukushima. Those drained soils are kept on the road side or in large flexible container bags. Sometimes, the soil is streamed out by rain water, which connects to a second damage to the nature. Thus, the excavation of soil is not always a good method, especially it is not safe and easily applicable in slopy mountain area. Moreover, sometimes workers may be in danger of exposure to radioactive materials while working in the contaminated area. An effective method to remove radioactive materials from the contaminated soil *in situ* is necessary.

Electrokinetic method is introduced to remove heavy metals from contaminated soil (Suzuki 2006, Miura 2015). In general, the electrokinetic method is used with two electrodes vertically set up inside the soil, while a DC voltage is applied in the electrodes. The process may not be effective in mountain areas as the effluent solution contains the removed heavy metals (including radioactive materials), which might cause a secondary contamination of the soil.

A new method of electrokinetic process has been developed to solve this problem. A flat cathode is set up on the surface of the contaminated soil, whereas the anode is installed inside the soil as shown in Fig. 1. This method is named as Flat Electrode Method Electrokinetic (FEM-EK) process. The details of the electrode can be found in other papers (Kishida 2017, Hatakeyama 2017). The objective is to remove the contaminated materials from the soil and lift them up to the cathode, where the cathode absorbs the metal ions in it. Then the cathode electrode will be bent and kept in storehouse for several years according to the half life of the radioactive materials absorbed in it. For that, the electrode must be flexible and flat in shape. At the same time, it must be strong enough to be in the soil during the experimental period. Moreover, it must be conductive to serve as an electrode. The position of the anode inside the soil will be decided according to the behaviour of the heavy metals/ radioactive metals that are responsible for the contamination of the soil. For Cs, the installation will be about 5 cm inside the soil as Cs from the previous observation, exists around 5 cm from the surface of the ground (IAEA 2005). Cs is a single valence metal which can easily be ionized by releasing an electron and can be attached to materials in the soil. This paper deals with the process of manufacturing of cathode electrode which will be used in FEM-EK process *in situ* to remove Cs from the contaminated soil.

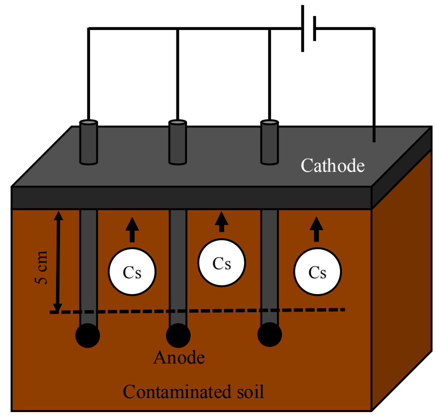


Fig. 1: Schematic diagram of FEM-EK process (Kishida 2017, Hatakeyama 2017).

MATERIALS AND METHODS

Materials: The conductivity of the electrode can be achieved from carbon powder. Burnt rice husks (Kuntan in Japanese) also possess the conductivity and there are some approaches to use burnt rice husks as electrodes in fuel cells (Kumagai 2009). In order to make the flat electrode, we have chosen natural materials that are also recyclable. Flat and flexible properties can be achieved by adding plant fibre cellulose or fibre type materials. Abaca is used in this experiment to get the flat type and flexibility of the electrode. Abaca is originally from Manila hemp; a tree quite similar to banana plant and whose biological name is *Musa textilis*. And finally, zeolite is added to the electrode for increasing of the adsorption of metallic ions. But zeolite is naturally a dielectric material which may affect the conductivity of the electrode.

Preparation methods: We followed the process quite similar to the nursery mat preparation introduced by Sanwa Tech Company, Tokyo, Japan. The electrode is made by using a suction device which works with a vacuum pump as seen in Fig. 2. The vacuum pump (G-50D, UlvacKiko Inc.) is connected with a glass bottle where the removed liquid from the electrode's material is kept. The mould for electrode is prepared with an acryl electrode fabricated frame. Plastic pipelines are connected with vacuum pump and glass bottle and electrode mould frame as shown in Fig. 2. The electrodes are made with three patterns of materials and processes. A brief description is provided below.

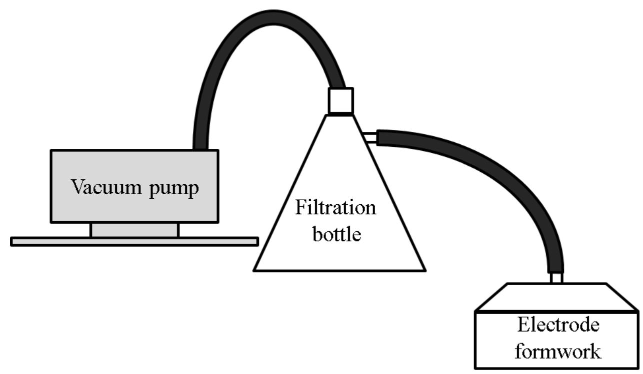


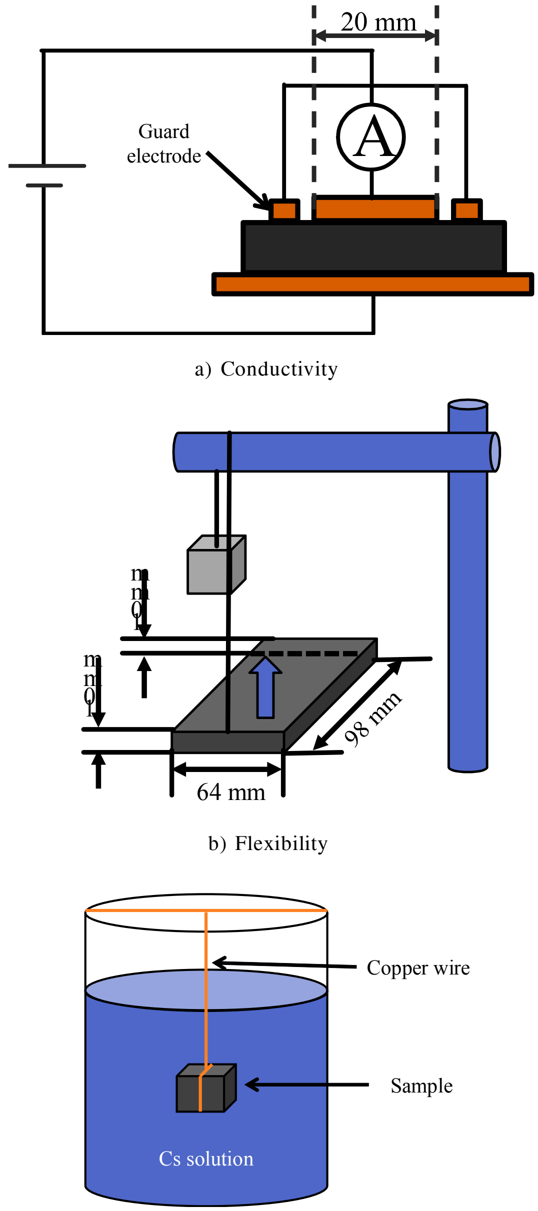
Fig. 2: Schematic diagram of electrode manufacturing apparatus (Hatakeyama 2017).

***Abaca and carbon powder*:** Carbon powders are good in conductivity and they are cheap. Fine carbon (grain size 5 mm) is used here. A 5.0 g of carbon powder and 5.0 g of abaca were used to make the electrode. However, as the grain size of the carbon is very small, it does not mix up well with abaca. We used 1.0 g of starch paste for mixing it well. The process is as follows:

1. 1.0 g of starch paste is inserted to 0.2 L of boiling water and stirred well by mechanical stirrer (SMT-102, AS ONE Co.), so that the starch paste can be melted well.
2. Abaca is placed in the bowl and then stirred by the same mechanical stirrer with 1000 rpm for 10 mins.
3. Then carbon powder of 5.0 g is added and stirred at 500 rpm for 10 mins.
4. The **suction** device method is used to prepare the shape of the electrode.
5. The electrode is dried at 60°C for 12 hours.

***Abaca and kuntan (burnt rice husks)*:** Kuntan is used instead of carbon powder in this method. Kuntan and abaca are mixed well and this time, no need of adding starch paste. The process is as follows:

1. 9.0 g of dried abaca is inserted to 0.2 L of water and then stirred by mechanical stirrer at 1000 rpm for 10 mins.
2. Then 1.0 g kuntan is added and stirred at 500 rpm for 10 mins.
3. The **suction** device method is used to prepare the shape of the electrode.



a) Conductivity

b) Flexibility

c) Cs adsorption

Fig. 3: Evaluation methods of the electrodes (Hatakeyama 2017).

4. The electrode is dried at 60°C for 12 hours.

Abaca, kuntan (burnt rice husks) and zeolite: Kuntan can adsorb Cs or heavy metals but at the same time Zeolite is also a well known Cs adsorber. To increase the Cs adsorption, we have tried another electrode by using kuntan, abaca and zeolite. The zeolite that we have used is a natural zeolite bought from Tohoku Zeolite Industrial Company. Zeolite is crushed by a blender machine and then the particles are run through a sieve and thus zeolite particles of 53 μm are chosen to prepare the electrode. Again, like the carbon particles, zeolite also does not mix up with abaca and kuntan due to its small size. So, starch paste is also added for this electrode. The process is as follows:

1. 1.0 g of starch paste is inserted to 0.2 L of boiling water and stirred well, so that the starch paste can be melted well.
2. Abaca is placed in the bowl and then stirred at 1000 rpm for 10 mins.
3. Then carbon powder 5.0 g is added and stirred at 500 rpm for 10 mins.
4. The **suction** device method is used to prepare the shape of the electrode.
5. The electrode is dried at 60°C for 12 hours.

EVALUATION OF THE ELECTRODE

Conductivity: Conductivity of the prepared electrodes is measured with a self made device as shown in Fig. 3. The device contains a circle of 20 mm of anode and a guard electrode to prevent the surface current from the measurement. A resistance meter (R6552, Advantest Co.) is used to measure the conductivity of the electrodes by measuring resistance with applied voltage. The conductivity is calculated from the basic knowledge of electric circuit.

Flexibility: Flexibility is one of the important characteristics of the prepared electrodes. If they are flexible then they can be settled on the vast area to remove the Cs. The flexibility is evaluated with the self made device as seen in Fig. 4. A side with 10 mm width of the electrode is kept still and the other side is tied with a rope which is connected with a weight ball of 70 g. The 100% flexibility is calculated when the rope can lift the electrode on a height of 88 mm (Fig. 4).

Adsorption of Cs: The electrodes are put in a 150 mL of non-radioactive Cs (^{133}Cs) solution for 1 day with a suspended Cu wire. For the observation, the electrodes are cut in to 20 mm×20 mm×20 mm and the test is performed. The Cs solution (20 ppm) was prepared from Wako Junyaku Company's standard Cs solution. The concentration of the solution is measured before and after the test with an ICP-MS (7500 Series, Agilent Technol.).

As the electrodes will be used *in situ*, and considering the Japanese weather condition, the conductivity and flexibility of the electrodes were measured in dry and wet conditions. To achieve the wet conditions, tap water was added from the centre of the electrodes during the measurement period.

RESULTS AND DISCUSSION

A snap shot is provided for kuntan (burnt rice husks) in Fig. 4. We have also observed the surface of the kuntan and abaca by a SEM (JSM-7800F, JEOL) (Figs. 5 and 6). Originally

Fig. 4: Snapshot of kuntan (burnt rice husks).

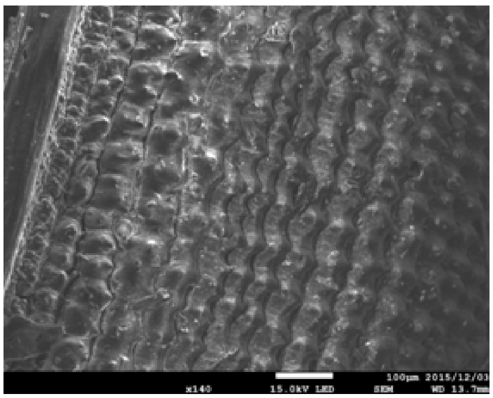


Fig. 5: Image of kuntan observed by SEM.

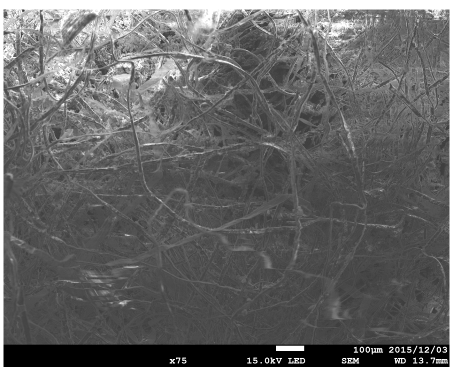
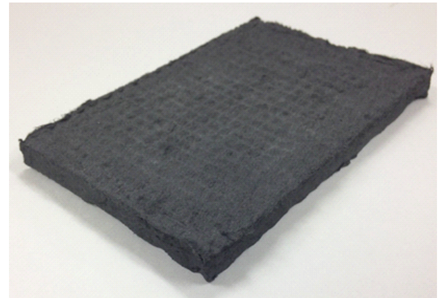


Fig. 6: Image of abaca observed by SEM.

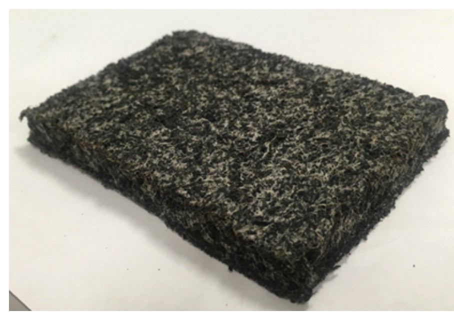
the rice husks are golden in colour but after burning they become charcoal in colour. The cellulose string like portions are showing the abaca which is essential for flexibility of the electrodes.

The three types of electrodes were prepared by using the methods described in previous section. The examples of the electrodes can be seen in Fig. 7. From the physical view and touching the electrodes, it can be understood that in order to achieve the flexibility of the electrodes, the mixing of abaca and conductive materials is very important in this method.



a) Carbon and abaca electrode

b) Kuntan and abaca electrode



c) Kuntan, abaca and zeolite electrode

Fig.7: Photo of the electrodes.

We have measured the conductivity and flexibility of the electrodes. The measurements are carried out in wet and dry condition of the electrodes. The electrodes will be used in practical field and they can be easily wet by rain, fog, mists and even water absorbed from the surface of the ground. So, as it is described earlier, the conductivity and flexibility of the electrodes were measured in dry and wet conditions. The conductivity of the electrodes at dry condition were found to be 10×10^{-6}, 0.8×10^{-6} and 0.04×10^{-6} S/m respectively. Carbon with abaca electrode showed the best performance in conductivity at dried condition. It proves again that carbon is a very good conductor. However, for the wet condition, the tap water dominates the conductivity and the values for the electrodes were found at around 800×10^{-6} S/m. Apart from conductivity, the electrode made from kuntan and abaca proved the best flexibility perform-

Table 1: Evaluation tests of the prepared electrodes.

properties		electrodes made for FEM-EK process		
		Carbon + Abaca	Kuntan + Abaca	Kuntan + Abaca + zeolite
conductivity [μS/m]	dry	10.0	0.80	0.04
	wet	800	740	800
flexibility [%]	dry	0.10	5.10	4.00
	wet	1.40	13.6	12.7
adsorption rate of ^{133}Cs	[%]		48.6	62.6
	[g/kg]		16.6	19.5

ance among the electrodes and the values for the electrodes were found around 800×10^{-6} S/m. Hence, the electrodes made from kuntan and abaca showed the best flexibility performance among the electrodes and the value was 5.1% at dry condition and 13.6% at wet condition. Among the three types of electrodes, the carbon electrode showed the lowest performance both in dry and wet conditions (i.e. 0.1% and 1.4%). The values can be found in Table 1.

The electrodes were prepared for using them in FEM-EK process where flexibility and conductivity are necessary conditions. However, from our measurements, it is clear that the carbon electrode has almost no flexibility. So, it can be understood that this carbon electrode has no merit in using it in FEM-EK process. Hence, the adsorption quality of the carbon and abaca electrode was not measured. For other two electrodes, the adsorption quality was measured. The piece of the kuntan and abaca electrode's adsorption rate of Cs was 48.6% (16.6 g/kg) in one day. Whereas, the electrode made with kuntan, abaca and zeolite proved more adsorbing ability of Cs with 62.6% (19.5 g/kg) from the solution. This difference is for the zeolite as zeolite is a well known absorber of Cs. These two electrodes can possibly be used in FEM-EK process.

CONCLUSION

In order to decontaminate the Cs from the polluted soil, we have developed a new electrokinetic method under the name of FEM-EK process. This method needs a special type of cathode electrode which is flexible, conductive and well adsorber of Cs. We have tried to use different conductive materials like kuntan, carbon and abaca for flexibility to prepare the electrodes. The electrodes made with kuntan and abaca showed good flexibility and conductivity. Again, the adsorption of Cs has increased with the addition of natural zeolite in the electrode though it harms in conductivity and flexibility. Our next step is to use these electrodes in FEM-EK process at lab scale and gradually at field to remove Cs from the polluted soil.

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