

Implication of Biological Activated Carbon Process Performance and Microbial Growth on Granular Activated Carbon for the Removal of Dissolved Organic Carbon in Water Purification

DO HUN LIM[†], YOON JIN LEE* and YOUNG SONG KO[‡]

*Department of Environmental Engineering, Cheongju University
36, Naedok-dong, Sangdang-gu, Cheongju, Chungbuk, 360-764, Republic of Korea
E-mail: yjlee@cju.ac.kr*

This research was performed to choose a suitable activated carbon when a biological activated carbon process was applied and the removal efficiency of organic carbon with biological activated carbon was assessed. Samples of different origin, such as river, lake water and humic acid, using four different types of activated carbon were investigated. The highest removal of dissolved organic carbon was obtained with Cargon and Norit activated carbon for a sample of Han river water and humic acid, respectively. Powder activated carbon appeared to be more efficient at removing dissolved organic carbon than was granular activated carbon for the sample of Han river water. Examination of the apparent molecular weight distribution showed that organic material over 10,000 daltons was removed, while low molecular weight distribution under 1,000 daltons increased after treatment with activated carbon of the sample of Han river water. The breakthrough point for Picabiol and Norit activated carbon was reached faster than was that for Cargon and Samchully activated carbon. An increasing order for biomass formation on activated carbon was shown for Cargon, Picabiol, Samchully and Norit activated carbon, respectively.

Key Words: Biological activated carbon, Granular activated carbon, Apparent molecular weight distribution, Dissolved organic carbon, Water purification.

INTRODUCTION

Surface water is a main source of raw water for producing supplied water in Korea. However, it is readily exposed to various forms of harmful pollution. Existing water treatment processes usually consist of a conventional system, which utilizes a combination of coagulation, filtration and chlorine disinfection. However, the desire of consumers for clean and reliable drinking water has increased with

[†]Donghae Engineering and Consultant Co., Ltd, Department of Environmental Engineering, Woorim e-Biz Center 610, 280-21 Seongsu-2-ga, Seongdonggu, Seoul 133-121, Republic of Korea.

[‡]HN Co., Ltd. Technical Institute of HN, Sihwa Industrial Complex 5 ba, Seonggok-dong, Danwon-gu, Ansan, Gyeonggi-do 429-936, Republic of Korea.

improvements in public standard of living level. More stringent standards for tap water are also being applied, so improvement of existing conventional purification systems or the development of substitute advanced water treatment facilities, is now required for a number of water utilities.

Biological activated carbon is a process which removes organic pollutants from water, both by adsorption reactions onto activated carbon and by biological decomposition by microorganisms¹⁻⁴. Because the level of microorganisms is comparatively low at the initial operation period, organic pollutants are mostly removed by the adsorption reactions and can be eliminated by microbial reaction as microorganisms grow on the surface of the activated carbon with the passage of time. The growth of microorganisms on activated carbon has the effect of regeneration of activated carbon and extension of the usage period of activated carbon⁵. The simultaneous reaction of bio-decomposition and adsorption in biological activated carbon process is reported to have a complicated interrelationship⁶.

Biological activated carbon process was applied to reduce microbial re-growth in water distribution, to reduce the formation of disinfection by-products and to remove micro-pollutants and taste and odor inducing materials⁷⁻¹¹. However, the selection of activated carbon is important for the introduction of biological activated carbon in water treatment process because that choice affects both the treatment efficiency and its economic feasibility^{10,12}.

The efficiency of the biological activated carbon process is dependent on the sort and mass level of microorganisms, because microorganisms are attached onto and inhabit the activated carbon and use natural organic matter in water as their substrates¹¹. The water influencing factors, such as temperature, kinds of organic matter and the quality of granular activated carbon, affect for the efficiency of the biological activated carbon process^{13,14}. Therefore, the analysis of biomass assay and microorganism composition gives important information for the optimal operation of biological activated carbon.

Natural organic matter is a complex body of organic matter, which has various sizes and chemical structures as a result of diverse mutual reactions. Organics that have molecular weight between 500 and 1,000 daltons are effectively removed by granular activated carbon, while high molecular weigh organics are not eliminated¹⁵⁻¹⁷. Kim *et al.*¹⁸ reported that the removal efficiency was shown 50-75 % for the organics over molecular weigh 11,000 daltons, when Nakdong river water was used as a water treatment sample after coagulation and sedimentation with 8-12 mg/L FeCl₃ dosage.

This research is performed to select an optimum activated carbon when a biological activated carbon process is installed for removal of organic matter by the reaction of biodegradation and adsorption reactions. Four different kinds of activated carbon, which had different material properties, were compared for their ability to remove organic matter from water samples of different origins based three samples, river, lake, humic acid. The behaviour of microorganisms in attaching and growing was observed during the operation time. Each biological activated carbon process

was assessed for the attached biomass and apparent molecular weight distribution of organic carbon. In addition, observations were made of surface variations of activated carbon by SEM following the application of the biological activated carbon process.

EXPERIMENTAL

Ilkam lake was used to raw water for operating the biological activated carbon columns. Raw water in a reservoir was pumped with an up-ward flow after it had passed through a sand filter. Columns manufactured from acrylic cylinders, had diameter 30 mm, height 250 mm and volume 120 mL. Columns were filled to a 170 mm height with activated carbon. Inflow rate was 10 mL/min and empty bed contact time (EBCT) was 12 min for these columns. A schematic diagram of the experimental equipment is presented in Fig. 1. The commercial granular activated carbon products, Cargon, Norit, Picabiol and Samchully, were used as the activated carbon sources and the physical properties of granular activated carbon used are presented Table-1.

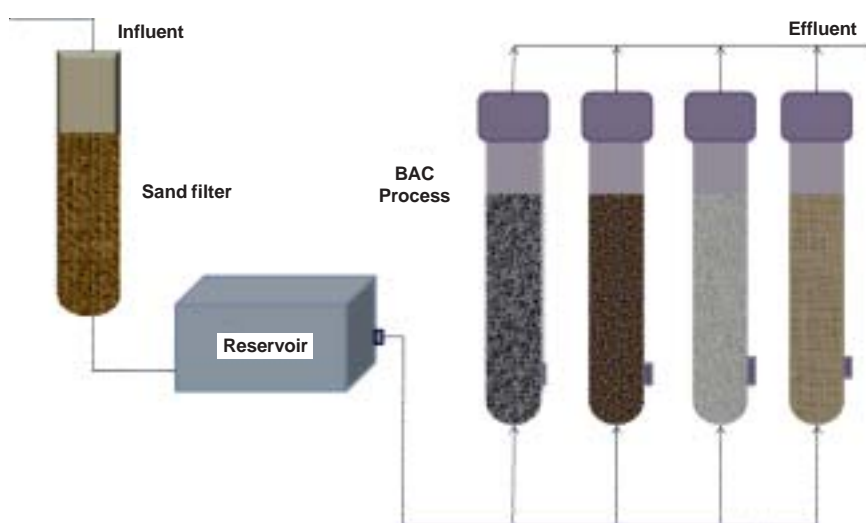


Fig. 1. Schematic diagram of biological activated carbon (BAC) treatment system

TABLE-1
PHYSICAL PROPERTIES OF GRANULAR ACTIVATED CARBON USED

Properties	Cargon	Norit	Picabiol	Samchully
Raw material	Bituminous coal	Peat	Lignite	Coconut
Apparent bulk density (g/mL)	0.43	0.38	0.24-0.3	0.43-0.48
Specific surface area (m ² /g)	1,050-1,200	1,200	1,200	1,100
Total pore volume (mL/g)	0.85-0.95	1.0	1.02	0.5-0.6
Ash content (%)	5	8	5	5
Moisture content (%)	0.9	2	7	5

Han river and Ilkam lake water were selected as natural water samples. Han river water was taken from a point under Chamsil Bridge as a river water sample. Ilkam lake, located on the campus of Konkuk University in Seoul, was sampled as a lake water sample. Samples were collected by grab sampling and were analyzed for pH, dissolved organic carbon, NH₃-N and HPC after they were brought into laboratory and the remainder stored under 4 °C. The source of humic acid was commercial product supplied by Aldrich Chemical. 1 g of humic acid was dissolved in a solution of 1 N HCl, then after, ultrapure water was added and the solution was agitated at the room temperature. The solution was then filtered through 47 mm diameter, 0.45 µm pore size membrane filters (Adventec) and stored at 4 °C.

Experimental procedures: The adsorption isotherm coefficients were obtained by the following process. Granular activated carbon was washed with distilled water until no further impurities were discharged. Granular activated carbon was then dried in a drying oven at a controlled temperature of 105 °C for 24 h. Then, it was shattered to a 200 mesh size, dried further 24 h at 105 °C and stored in a desiccator. 100 mL of Han river, Ilkam lake or humic acid solution was transferred to 1 L flasks and 0.5, 1, 2, 5, 10, 20, 30, 40, 50 and 100 mg of powder activated carbon was added, followed by with the agitation at 200 rpm. Samples for the adsorption experiment were taken after 4 h and filtered through 0.45 µm membranes pre-washed with 150 mL of distilled water.

An experiment was performed for the Han river, Ilkam lake and Aldrich humic acid solutions to compare of the molecular weight distribution before and after the adsorption reaction. 0.05 g and 0.2 g granular activated carbon and powder activated carbon were added to 500 mL samples and agitated at 200 rpm for 4 h. Samples were then left for 2 h to allow sedimentation of activated carbon. Molecular weight distribution was analyzed after filtration through a 0.45 µm membrane. Microbial oxygen consumption was measured with a respirometer. Activated carbon with attached microorganisms was taken from sampling ports of each column and incubated in 10, 20 and 30 mg/L glucose solutions.

Surface specimens of activated carbon were observed by scanning electron microscope (SEM) analysis. 0.1 g of activated carbon was washed and transferred to a phosphate buffered solution of glutaraldehyde solution and stored in a refrigerator for 4 h. Activated carbon samples were then fixed in 1 % osmium tetroxide for 0.5 h and washed with phosphate buffer solution. After a final wash with ethyl alcohol, samples were stored in desiccator. Activated carbon pretreated by this method was fixed to a cell strut and coated with platinum. Photography was done using a Leika Stereo Scan 440.

Analytical methods: The molecular weight distribution was analyzed by ultra-filtration (Amicon 8200) which consisted of 20 mL volumes of UF cells and clamps. Samples were filtered through by applying 55 psi of pure nitrogen and were stirred by a magnetic stirrer to reduce intensity polarization. After filtration through membranes, samples were collected in vials and were treated with acid and heat. The initial 10 mL

of the samples was from the filtration discarded by membrane washing to reduce differences and the last 90 % was used to measure dissolved organic carbon. Data were recorded as low molecular weight (LMW), medium molecular weight (MMW) and high molecular weight (HMW) for the molecular weight ranges < 1,000, 1,000-10,000, >10,000 daltons, respectively, for convenience in this study.

The assay for phospholipid analysis was performed using the method of Findlay *et al.*¹⁹. Activated carbon was sampled from the sampling port located at the lower part of the columns. Activated carbon samples were washed with distilled water and the extraction was performed on 2 g activated carbon. This analysis was performed immediately after washing the activated carbon because phospholipids are easily decomposed. A chloroform: methanol: phosphate buffer (1:2:0.8) was added to the activated carbon samples in 70 mL vials, followed by strong agitation. The suspensions were left standing for 2 h, then, 7.5 mL of chloroform and distilled water were added, the samples were agitated again and left standing a further 24 h. The upper water layer was removed and the lower chloroform layer was collected and filtered through a GF/C membrane. The chloroform phase containing the lipids was evaporated under a stream of nitrogen. The dried phospholipids were digested by adding 0.45 mL of a saturated potassium persulfate solution and heating the ampoules for 2 h at 100 °C. 0.2 mL of ammonium molybdate was added to react with the liberated phosphate and the mixture was allowed to stand for 10 min. The phosphomolybdate was complexed with 0.45 mL malachite green and let stand for 0.5 h. The absorbance at 610 nm was then measured for each sample using a UV-visible spectrophotometer (UV-1601, Shimadzu). The assay data were expressed as nmoles phosphate/gram of media.

HPC was inoculated by spreading a plate on an R₂A medium, incubating for 7 d at 20 ± 1 °C. Data were expressed as colony forming units (CFU)/mL. Dissolved organic carbon was analyzed by a TOC 500 analyzer (Shimadzu) in NPOC mode. UV254 was measured using an UV visible spectrophotometer (UV 1601, Shimadzu) with a 10 mm cell at 254 nm.

RESULTS AND DISCUSSION

Dissolved organic carbon adsorption characteristics: The coefficients, $1/n$ and K of Freundlich isotherms for Han river water is shown in Table-2. The K value, which represents the extent of adsorption capability, was found to be 20.5, 12.7, 12.7, 1.63 for Cargon, Norit, Picabiol, Samchully activated carbon, respectively. The K value was evidently low for Samchully activated carbon. Cargon activated carbon was shown to have the highest K value of the selected activated carbon. Therefore, it was evaluated to have highest adsorption ability. Jung *et al.*²⁰ reported $1/n$ and K values of 0.047 and 2.20 for Nakdong river water. The value of K for raw water of Chungju and Puyo water treatment facilities after treatment of domestically produced powder activated carbon was estimated at 20.88 and 10.05, respectively²¹.

TABLE-2
EXPERIMENTAL VALUE OF THE FRUNDLICH ISOTHERM PARAMETNER FOR
HUMIC ACID, ILKAM LAKE AND HAN RIVER

Sample	Cargon		Norit		Picabiol		Samchully	
	K	1/n	K	1/n	K	1/n	K	1/n
Humic acid	2.548	3.597	5.962	2.465	5.574	2.689	1.923	3.813
Ilkam lake	12.11	5.282	1.704	6.691	74.81	4.826	4.669	4.412
Han river	20.46	1.593	12.74	2.354	12.73	2.363	1.634	2.078

The order of K value for humic acid from highest to lowest was found with Norit, Picabiol, Cargon, Samchully activated carbon, respectively. Norit activated carbon was shown to have the best adsorption function for humic acid of the selected activated carbon. The K coefficient for Norit was 3.1 times higher than that of Samchully activated carbon.

The value of 1/n was between 4.40 and 6.69 for all four types activated carbon for Ilkam lake sample. The values of 1/n for the lake water were far higher than those of the other water samples. The lowest value of 1/n was found for Samchully activated carbon. This means that the Samchully activated carbon had the highest affinity for organic materials in the Ilkam lake water, followed by Picabiol, Cargon and then Norit activated carbon. The K value was found to be highest for Picabiol activated carbon, which was 44 times higher than that of Norit. The K values, in order from highest to lowest, were shown to be Picabiol, Cargon, Samchully and then Norit.

Dissolved organic carbon removal after activated carbon adsorption on different activated carbon types with the samples from Han river, Ilkam lake and humic acid are shown in Fig. 2. Han river water treated with 400 mg/L of granular activated carbon and powder activated carbon. Powder activated carbon was shown to have a better dissolved organic carbon removal efficiency than that of granular activated carbon with the treatment of Han river water. The difference was largest for the Picabiol activated carbon. The dissolved organic carbon removal efficiency for Han river sample was similar for Picabiol, Cargon and Norit powder activated carbon with a mean value of 66 %. The order of dissolved organic carbon removal from highest to lowest was shown to be Cargon, Norit, Picabiol and then Samchully for the 400 mg/L granular activated carbon treatment of the Han river water.

Dissolved organic carbon removal did not show great difference on activated carbon sorts for the treatment of sample of Ilkam lake. Samchully activated carbon was found to have the lowest absorption effect for dissolved organic carbon removal for lake water. Picabiol, Cargon and Norit activated carbon were shown to have similar dissolved organic carbon removal efficiency and the mean removal of dissolved organic carbon was 46 and 64 % for 100 and 400 mg/L of activated carbon, respectively, for the samples of Ilkam lake.

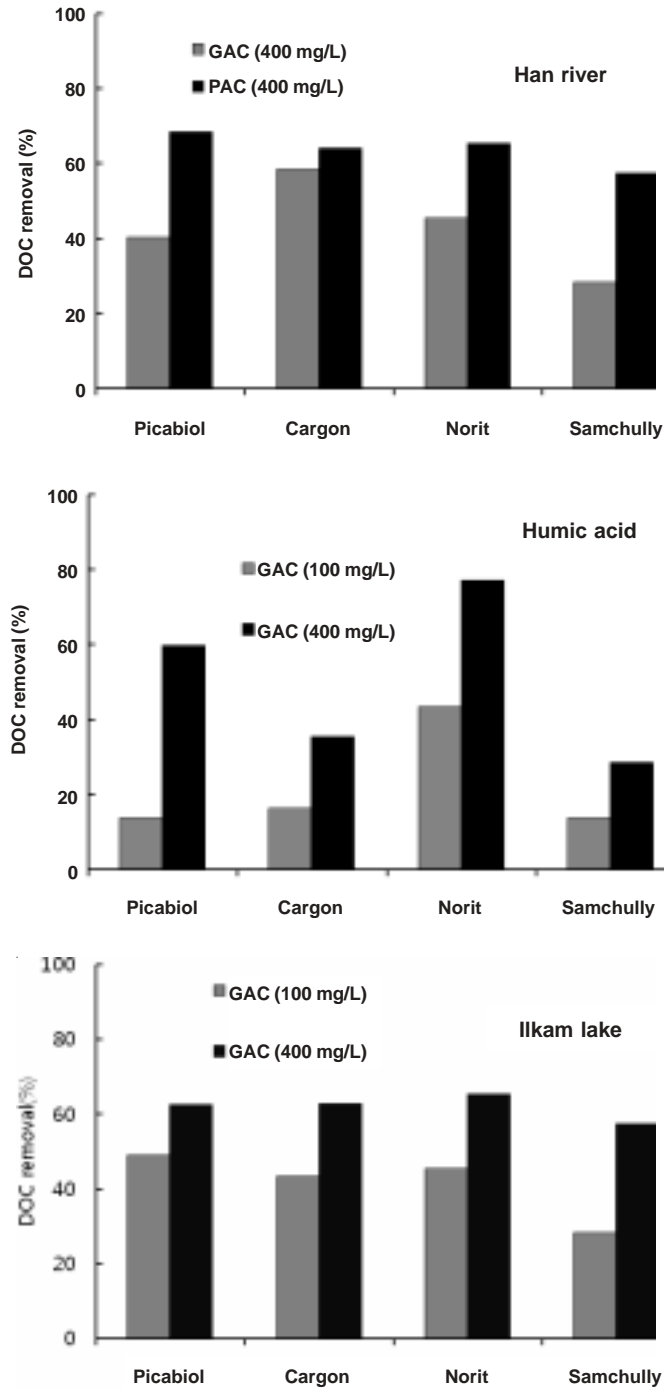


Fig. 2. Dissolved organic carbon (DOC) variation after application of Picabiol, Cargon, Norit and Samchully activated carbon with Han river, humic acid and Ilkam lake

The best removal of dissolved organic carbon in humic acid solutions occurred after treatment with Norit activated carbon. However, the level of dissolved organic carbon removal efficiency was similar for 100 mg/L of Picabiol, Cargon, Samchully activated carbon. Dissolved organic carbon removal was increased with granular activated carbon dose. The order of dissolved organic carbon removal from highest to lowest for humic acid samples was Norit, Picabiol, Cargon and then Samchully activated carbon with the application of 400 mg/L.

The variation in molecular weight distribution after the application of granular activated carbon (400 mg/L) and powder activated carbon (400 mg/L) for Han river water is presented in Fig. 3. The molecular weight distribution of organic matter of under 1,000 daltons, between 1,000 and 10,000 daltons, over 10,000 was shown to 32, 14, 54 %, respectively, in this experiment for the Han river water. The highest distribution for Han river water was shown in the weight range over 10,000 daltons. In the range of > 10,000 daltons, 50 % of the weight distribution was over 30,000 daltons. Bae²¹ reported a molecular weight distribution of 16, 21 and 63 % for the molecular weight of under 1,000 daltons, between 1,000 and 10,000 daltons, over 10,000 daltons, respectively, for raw water from the Cheongju water treatment utility.

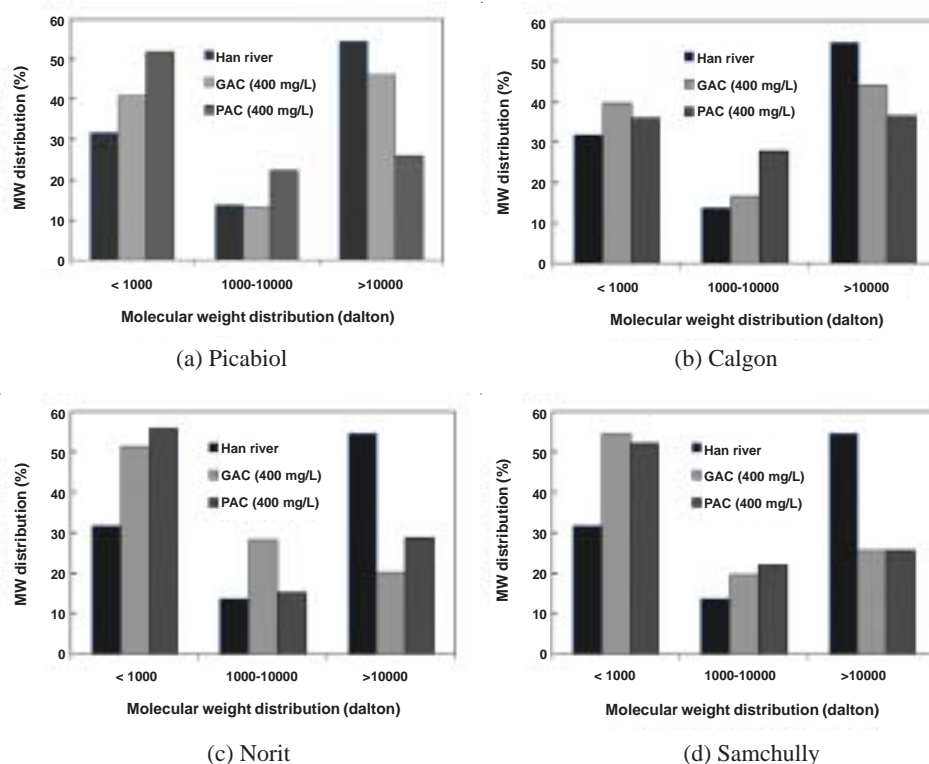


Fig. 3. Variation of molecular weight (MW) distribution after granular activated carbon (GAC) and powder activated carbon (PAC) adsorption treatment of Han river water

Powder activated carbon appeared to have better removal efficiency for dissolved organic carbon than did granular activated carbon for the treatment of Han river water. That might have been caused by powder activated carbon having more adsorption sites because powder activated carbon has more surface area than granular activated carbon. There was no great difference in dissolved organic carbon removal efficiency among the different types of powder activated carbon for Han river water. However, Picabiol powder activated carbon was still shown to have the highest dissolved organic carbon removal efficiency of the materials tested.

Molecular weight distribution of dissolved organic carbon for Ilkam lake water was found to be 35, 26 and 39 % for under 1,000 daltons, between 1,000 and 10,000 daltons, over 10,000 daltons, respectively (Fig. 4). After granular activated carbon adsorption, the highest removal was appeared for the materials with molecular weight between 1,000 and 10,000 daltons.

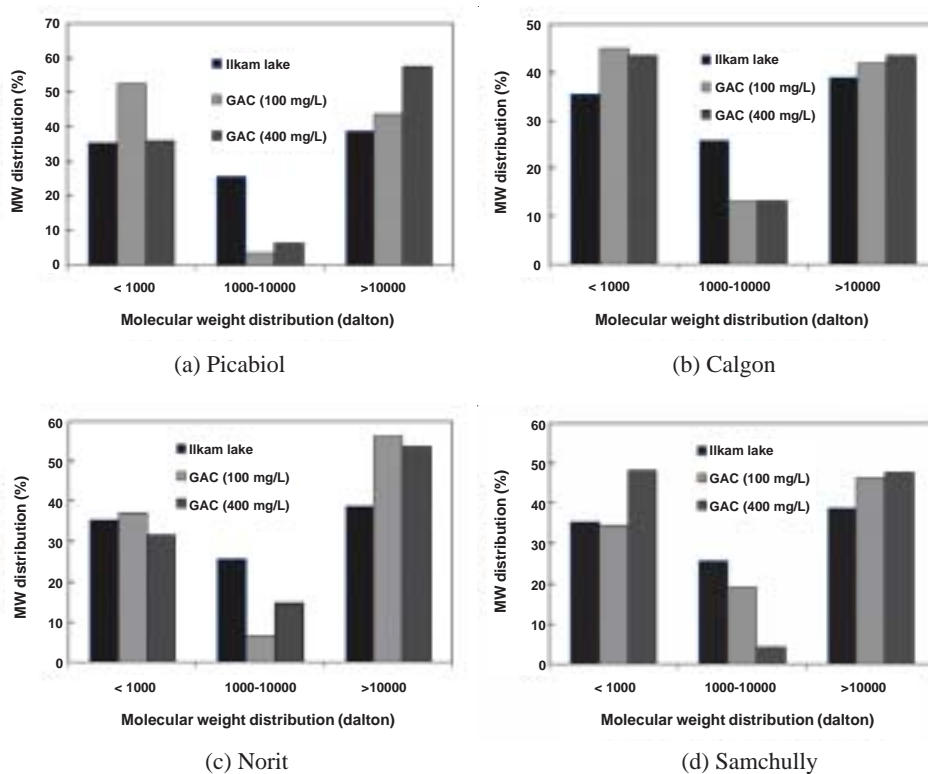


Fig. 4. Variation of molecular weight distribution after granular activated carbon (GAC) and powder activated carbon (PAC) adsorption treatment of Ilkam lake water

Dissolved organic carbon distribution in humic acid was analyzed as 7, 20 and 73 %, respectively for molecular weight of under 1,000 daltons, between 1,000 and 10,000 daltons and over 10,000 daltons (Fig. 5). The efficiency of dissolved organic

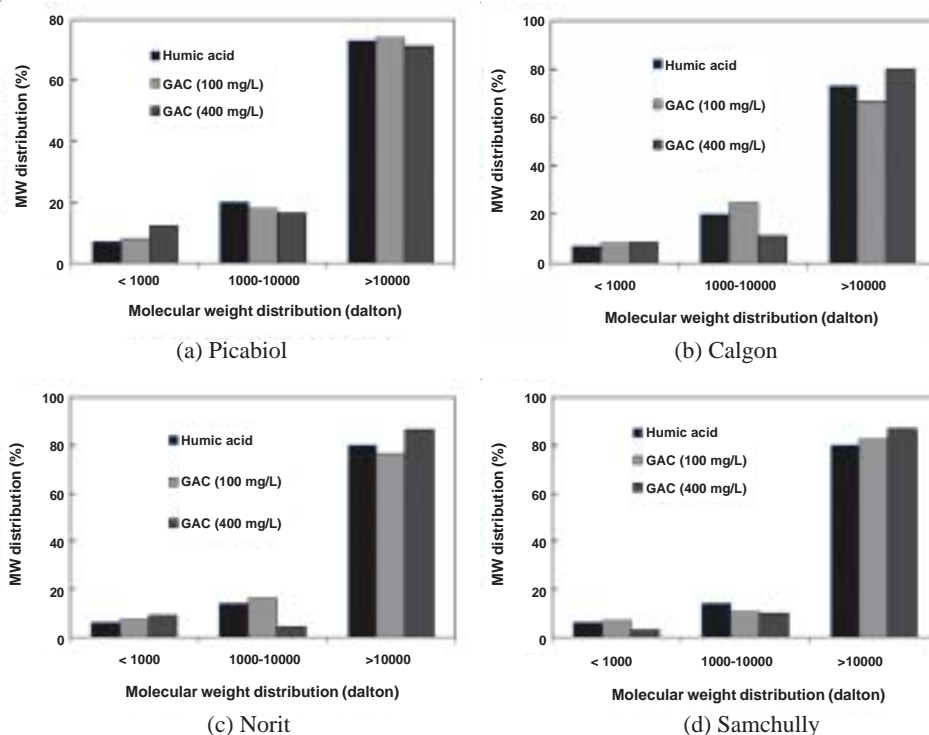


Fig. 5. Variation of molecular weight distribution after granular activated carbon (GAC) adsorption treatment of humic acid

carbon removal was low for material of molecular weight over 10,000 daltons, which composed of largest proportion of the humic acid sample. This result was consistent with Choi *et al.*²² which found that the variation of molecular weight over 10,000 was not noticeable in a solution made from Aldrich humic acid following the treatment with activated carbon while molecular weight over 3,000 increased after activated carbon adsorption process.

The removal efficiency for the molecular weight distribution between 1,000 and 10,000 daltons was 18, 43, 69 and 28 % after adsorption with 400 mg/L activated carbon of Picabiol, Cargon, Norit and Samchully, respectively. Norit activated carbon was found to have the highest removal efficiency for the dissolved organic carbon of molecular weight between 1,000 and 10,000 daltons for humic acid samples.

Dissolved organic carbon removal: The level of dissolved organic carbon in influent and effluent varied with bed volume of each column, as shown in Fig. 6. The initial ratio of the dissolved organic carbon in the effluent to the dissolved organic carbon in the influent was 0.39, 0.41, 0.33 and 0.55 for Cargon, Norit, Picabiol and Samchully activated carbon, respectively. Samchully activated carbon was shown to have the highest initial value of the selected activated carbon types. The highest value appeared in the bed volume range between 1,000 and 1,500 for

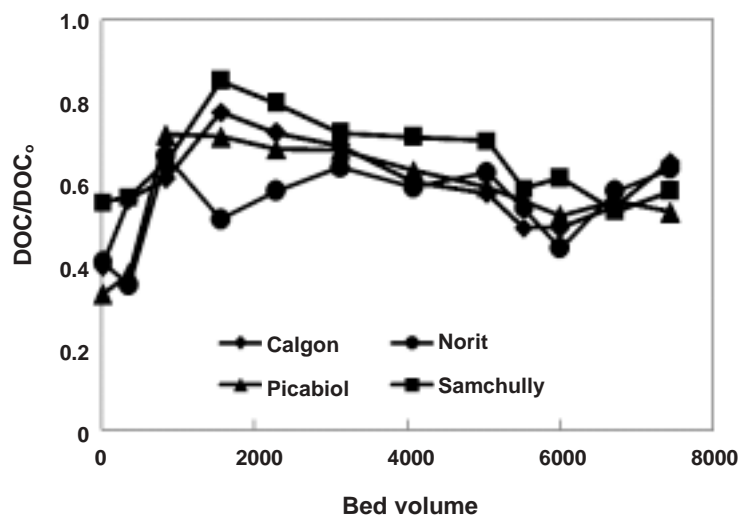


Fig. 6. Variation of ratio of dissolved organic carbon effluent (DOC) and dissolved organic carbon influent on bed volume (DOC_0)

the selected activated carbon types used in this experiment. The removal of dissolved organic carbon was attained by adsorption of activated carbon itself initially, until the bed volume reached the range between 1,000 and 1,500, at which point the activated carbon reached the limit of its adsorption ability. The decrease in dissolved organic carbon after the breakthrough might have been due to microorganisms attached on activated carbon.

The removal rate of organic matter was found to be highest for the Norit column. Adsorption capacity for Samchully activated carbon was relatively lower than that of the others. Organic removal rate by microorganisms in the Norit column was shown to be *ca.* 10 to 20 % higher than that of the Samchully column.

The ratio of dissolved organic carbon in effluent and dissolved organic carbon in influent was shown to have the highest value with a bed volume of 1,000 for Picabiol and Norit activated carbon and with a bed volume of 1,500 for Samchully and Cargon. This means that the use of Picabiol and Norit activated carbon reaches the point of breakthrough earlier than does Samchully and Cargon activated carbon. The value of dissolved organic carbon ratio between influent and effluent was stabilized between 0.5 and 0.6 at bed volume of 1,500 and it decreased until a bed volume of 6,000. Lee and Kim² reported that microbial activation occurred after 4 month operation of an ozone-granular activated carbon process.

Level of attached microorganisms: The level of microorganisms attached to the activated carbon for a bed volume of 4,080 and 6,500, measured by phospholipids analysis, is shown in Fig. 7. The cell numbers per activated carbon unit gram were 1.05×10^9 , 0.87×10^9 , 0.87×10^9 , 0.79×10^9 for Cargon, Norit, Picabiol and Samchully activated carbon, respectively, for a bed volume of 4,080.

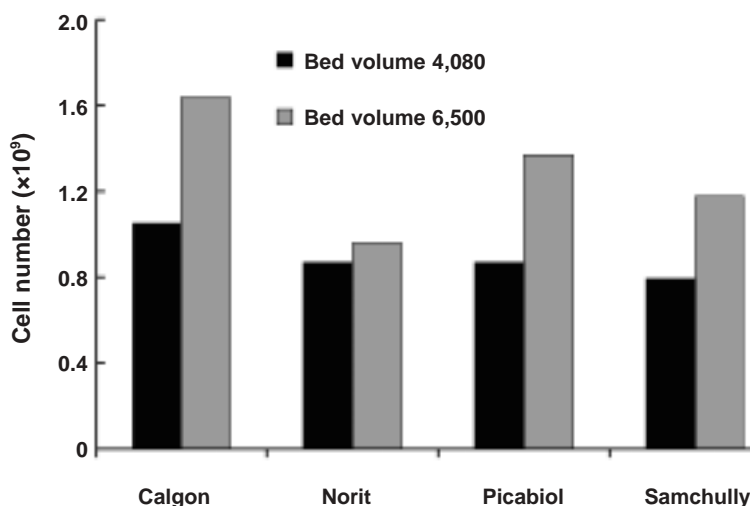


Fig. 7. The level of cell for the different sorts of activated carbon at the bed volume of 4,080 and 6,500

The cell number per activated carbon gram was found to be 1.64×10^9 , 0.96×10^9 , 1.37×10^9 , 1.18×10^9 for Cargon, Norit, Picabiol and Samchully activated carbon, respectively, for a bed volume of 6,500. Cargon activated carbon was shown to have the highest cell number at both bed volumes. Cell number increased with a range of 10 to 57 % with the bed volume increase from 4,080 to 6,500. The cell number on Norit activated carbon was shown to have the lowest value at a bed volume of 6,500. Organic removal (Fig. 6) increased with the increase in cell level except for Samchully activated carbon. This tendency was more distinct for the Cargon and Picabiol activated carbon.

Oxygen consumption was measured to estimate the vitality level of microorganisms, using glucose at 10 and 20 mg/L (Fig. 8). For glucose 10 mg/L, oxygen consumption for Samchully activated carbon was highest and it continuously increased as time passed. The activity level for microorganisms was comparatively low for Cargon activated carbon even though it had the highest cell numbers. The oxygen consumption of Cargon and Picabiol activated carbon were comparatively high, while Norit activated carbon had the lowest value at 20 mg/L glucose. The consumption of oxygen was lowest for Norit activated carbon, which also had the lowest value for cell number.

SEM observation was performed to evaluate the status of attached microorganism with activated carbon taken from each column (Figs. 9 and 10). Virgin carbon for Cargon and Samchully activated carbon appeared to be rough and macro-pores were distributed over the surface. However, virgin carbon of Picabiol and Norit showed mainly microporous surfaces.

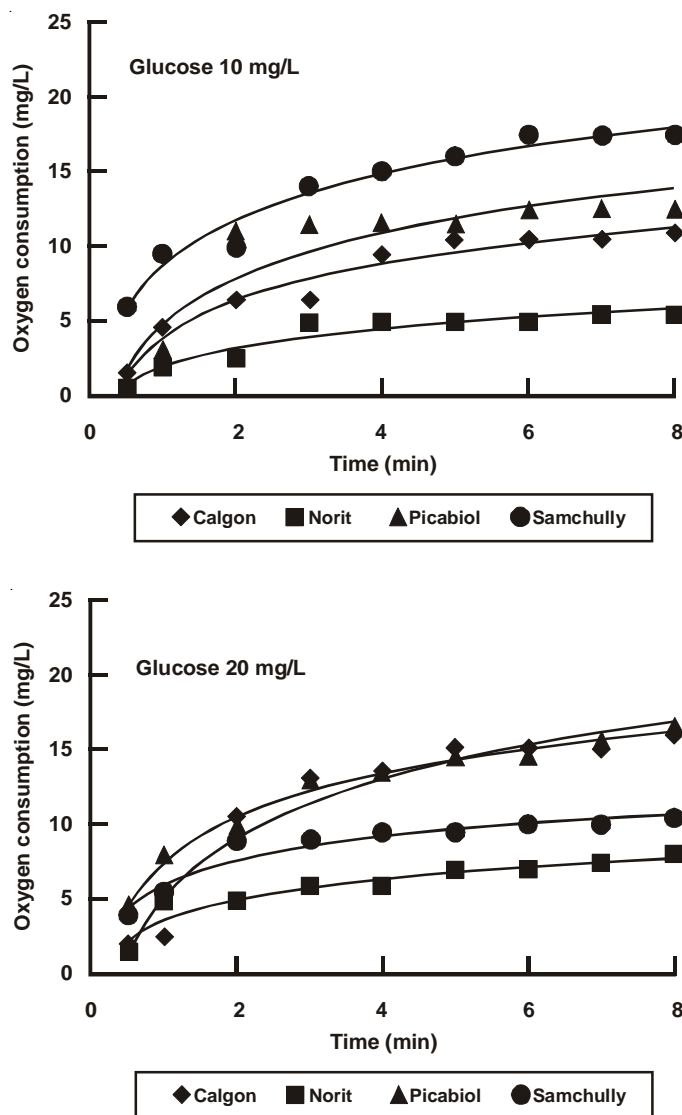


Fig. 8. Oxygen consumption of attached microorganisms on different activated carbon with 10 and 20 mg/L of glucose

The surface of Picabiol activated carbon was covered with a biofilm that looked like a number of dots entangled with each other. Microorganisms were seated on the pores. The surface of Norit activated carbon also showed mass of biofilm, which looked like the interwoven threads on the surface of the activated carbon pores. The dense biofilm layer was observed on the surface of Norit activated carbon. Cargon activated carbon was covered with features of filaments of microorganisms. The surface of the Samchully activated carbon column was found to have sporadic holes.

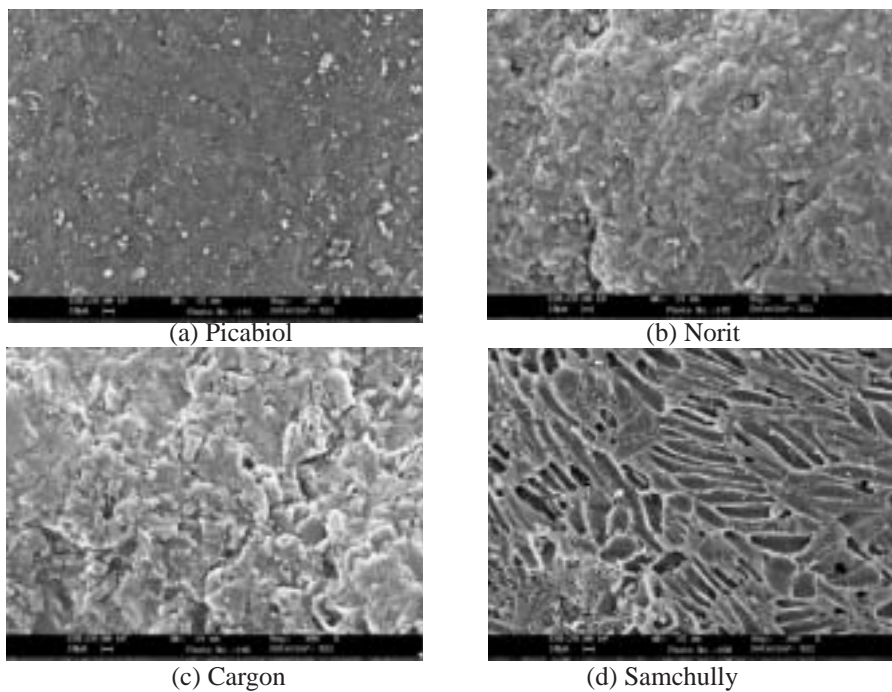


Fig. 9. SEM observations for the surface of virgin activated carbons

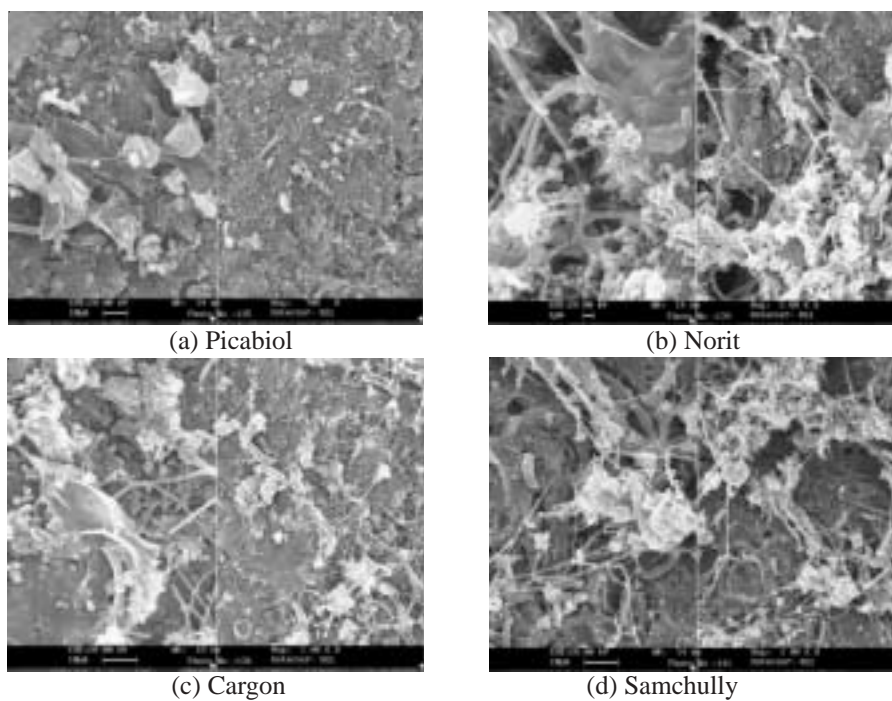


Fig. 10. SEM observations for the surface of activated carbons
(The right: 1,000 magnifications and the left: 3,000 magnifications)

Conclusion

The aim of this research was to evaluate the adsorption characteristics, variation of molecular weight distribution after adsorption, microbial distribution and vitality of attached microorganisms on different types of activated carbon available for sale on the domestic market.

Isotherms adsorption coefficients were shown to be highest for Picabiol and Cargon activated carbon for the treatment of Ilkam lake and Han river, respectively. Highest dissolved organic carbon removal efficiency was shown for Cargon and Norit for the activated carbon adsorption treatment of river water and humic acid. There was no notable difference between activated carbon types for their treatment of Ilkam Lake water. The efficiency of powder activated carbon was better than that of granular activated carbon for the treatment of Han river water. The distribution of HMW over 10,000 daltons decreased and the molecular weight area under 1,000 increased, after activated carbon adsorption of samples of Han river water. MMW between 1,000 and 10,000 daltons decreased and the molecular weight area under 1,000 daltons increased after activated carbon adsorption treatment for Ilkam lake water.

The time to reach the breakthrough point for adsorption for Picabiol and Norit activated carbon was faster than that for either Cargon and Samchully activated carbon. This means that application of Cargon and Samchully activated carbon are more beneficial than are Picabiol or Norit for the purpose of granular activated carbon adsorption. The removal of organic carbon was highest for Norit activated carbon.

The highest cell number, as estimated by phospholipids measurement, was found to be 1.64×10^9 cells/g for Cargon activated carbon. Biomass formed on activated carbon was higher in the order of Cargon, Picabiol, Samchully and Norit activated carbon.

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