A Study of Sorghum Distillery Residue Activated Carbon for Water Purification

Lin, Han Chien
Laboratory of Environment Functional Materials, Department of Wood Based Materials and Design, College of Agriculture, National Chiayi University

Hu, Jhih-Siang
Graduate Institute of Wood Based Materials and Design, College of Agriculture, National Chiayi University : Master's Program

Lee, Wen-Ju
Graduate Institute of Wood Based Materials and Design, College of Agriculture, National Chiayi University : Master's Program

Lai, Ying-Jang
Department of Food Science, College of Science and Engineering, National Quemoy University

他

KYUSHU UNIVERSITY

A Study of Sorghum Distillery Residue Activated Carbon for Water Purification

Han Chien LIN[1]*, Jhih–Siang HU[2], Wen–Ju LEE[2], Ying–Jang LAI[3], She–Ching WU[4] and Noboru FUJIMOTO

Laboratory of Wood Material Technology, Division of Sustainable Bioresources Science,
Department of Agro–environmental Sciences, Faculty of Agriculture,
Kyushu University, Fukuoka 812–8581, Japan
(Received May 8, 2015 and accepted May 19, 2015)

This study used fermentation waste–Sorghum Distillery Residue (SDR) as a precursor to prepare different kinds of Sorghum Distillery Residue Activated Carbon (SDRAC) by physical activation with steam, in order to evaluate the water purification and the preliminary safety of water quality before and after purification. The turbidity, total hardness, nitrite nitrogen, total bacterial count, and coliform of the water purified by prepared SDRAC were 0.12 NTU, 55.6 mg/L, 0.00 mg/L, 1 CFU/mL, and below 1.0 CFU/mL, respectively, meeting the quality standard for drinking water in Taiwan. The total organic carbon in water was reduced by 34.5%, at most. The cytotoxicity test results of the Ames Tests for the water specimens before and after filtration by SDRAC showed that the bacterial survival rate was higher than 80 % of control group, meaning that there is no cytotoxicity. The mutagenicity results showed that the spontaneous revertants were not exceeded by over two times, suggesting no mutagenicity. The SDRAC; therefore, has functional value of water purification, and can be preliminarily regarded as a safe natural water purifying material.

Key words: Sorghum Distillery Residue Activated Carbon (SDRAC), Physical Activation with Steam, Water Purification, Ames Tests

INTRODUCTION

Activated carbon is a porous adsorbing material with nonpolar surface and is effective in adsorbing organic matter from water solution (Tomaszewska and Mozia, 2002; Przepiorski, 2006; Villacan *et al*., 2006). It has stable chemical properties, and is resistant to acid, alkali, high temperature, and high pressure, and is extensively applicable to the drinking water purification process (Jiang, 2010). As activated carbon has a developed pore structure, it can adsorb the trace amount of organic contaminants in water (Kihn *et al*., 2000; Andersson *et al*., 2001), and can significantly reduce the chemical oxygen demand and total organic carbon in water (Kunio *et al*., 2001; Seyed *et al*., 2004; Omri *et al*., 2013). It is effective on the turbidity and chromaticity of the physical standards for drinking water (Anu *et al*., 2006). In addition, the activated carbon has inhibitory effect on total bacterial count and coligroup in water (Ogawa *et al*., 2011), and it conforms to the biological criteria of water standards of the Environmental Protection Administration of Taiwan (EPA). It is obvious that activated carbon is important for water purification. A previous study (Lin *et al*., 2015) reported that the Sorghum Distillery Residue

(SDR) is a type of fermentation waste, which can be prepared into activated carbon with multiple mesopores, and used as functional water purifying material. In Taiwan, tap water is from groundwater, reservoirs, and rivers (Shiu, 2000), and all water sources must conform to the Water Quality Standard for Drinking Water Sources (EPA, 1997), before it can be used as a water source of tap water. The water has to be purified by waterworks and conform to the water quality standard for drinking water, as specified by the EPA of Executive Yuan (2013c), before it is supplied as domestic water for the public.

In modern life, the massive amounts of chemical substances, environmental accumulation, biological concentration, biotransformation, and chemical reaction pollutes the environment and causes hazards to human health, such as malignant tumors, impaired capacity to bear children, and gene mutation. Mutagenicity is a change in the hereditary property in the reproduction process of deoxyribonucleic acid storing gene information in biological somatocyte caused by toxic chemical substances. If there is biological mutagenicity, there is a high proportion with biological carcinogenicity. Therefore, if the raw water of tap water is severely polluted, there may be mutagenicity increasing the probability of cell mutation. If the chlorine dosage is too high in the tap water treatment process, there may be high carcinogenic risk. In addition, activated carbon reduces the mutagenicity of water to different extents (Monarca *et al*., 1983; Marguerite *et al*., 1986). Therefore, gene mutation caused by chemical substances can be tested by the Ames Tests, in order to evaluate the carcinogenecity to the human body (Ames *et al*., 1975; Maron and Maron, 1983).

This study attempted to use the SDRAC prepared by physical activation in a previous study (Lin *et al*., 2015) to evaluate its application to water purification, as well

---

[1] Laboratory of Environment Functional Materials, Department of Wood Based Materials and Design, College of Agriculture, National Chiayi University, Chiayi, Taiwan, ROC
[2] Master, Graduate Institute of Wood Based Materials and Design, College of Agriculture, National Chiayi University, Chiayi, Taiwan, ROC
[3] Department of Food Science, College of Science and Engineering, National Quemoy University, Kinmen, Taiwan ROC
[4] Department of Food Science, College of Life Sciences, National Chiayi University, Chiayi, Taiwan, ROC
* Corresponding author (E–mail: alexhlin@mail.ncyu.edu.tw)

as the preliminary safety of water quality before and after purification. To reduce the economic cost of SDRAC, and use it locally, some reports indicate that the Kinmen County dam spring has high turbidity and high density coligroup, and the eutrophication of the reservoir increases the total organic carbon in water (Construction and Planning Agency, 2005). Therefore, this study used prepared SDRAC for Kinmen County water specimen purifying treatment and related water testing. The water specimen treated by the prepared activated carbon was used to evaluate the preliminary safety of cytotoxicity and mutagenicity of the Ames Tests. The activated carbon prepared by SDR is expected to conform to sustainable recycling of Kinmen distillery residue waste resource in order to purify local water, and such treated water is safe.

MATERIALS AND METHODS

Specimen

Sorghum Distillery Residue Activated Carbon (SDRAC)

The fermentation waste, Sorghum Distillery Residue (SDR), was used to prepare different kinds of SDRAC by physical activation with steam, and that with a yield above 17% and iodine value higher than 700 mg/g (T850–60–090–H_2O, T800–60–090–H_2O, T800–60–120–H_2O and T800–60–150–H_2O) were selected for water testing. This

study referred to Lin *et al.* (2015) for the codes, basic properties, and adsorption characteristics of SDRAC.

Source of water specimen

The raw water from the water source of the Kinmen County Ronghu purification plant, and the water specimen in front of the activated carbon filter (slow filter) following the water purification procedure of the Ronghu purification plant were used as the blank group (Blank). The raw water was obtained on May 27, 2014, and the slow filter water was obtained on May 27, 2014.

Methods

Water purification methods

Standing method: the SDRAC and the water specimens of raw water or slow filter water were mixed by a weight ratio of 1: 10 (wt%), and kept still for 30, 60, and 120 min, respectively (Zhou and Lu, 2010), before water testing;

Filtration method: the SDRAC was placed in a glass funnel, the raw water or slow filter water flowed from the top to the bottom under gravity, and were controlled by valve; the flow velocity was 10±2 mL/min and 5±2 mL/min (Ogawa *et al.*, 2011), and the weight ratio of SDRAC to raw water or slow filter water was 1: 10 (wt%). The abbreviations for water purification specimens are as shown in Table 1.

Table 1. Abbreviations of water specimen after processing by water purification with SDRAC[1]

Type of SDRAC [2]	Method of water purification	Time of standing and flow velocity	Water specimen
T850–60– 90–H_2O [3]	Standing Method	30 min	T850–60–090–030min [4]
		60 min	T850–60–090–060min
		120 min	T850–60–090–120min
	Filtration Method	10 mL/min	T850–60–090–10 mL/min
		5 mL/min	T850–60–090–05 mL/min
T800–60– 90–H_2O	Standing Method	30 min	T800–60–090–030min
		60 min	T800–60–090–060min
		120 min	T800–60–090–120min
	Filtration Method	10 mL/min	T800–60–090–10 mL/min
		5 mL/min	T800–60–090–05 mL/min
T800–60–120–H_2O	Standing Method	30 min	T800–60–120–030min
		60 min	T800–60–120–060min
		120 min	T800–60–120–120min
	Filtration Method	10 mL/min	T800–60–120–10 mL/min
		5 mL/min	T800–60–120–05 mL/min
T800–60–150–H2O	Standing Method	30 min	T800–60–150–030min
		60 min	T800–60–150–060min
		120 min	T800–60–150–120min
	Filtration Method	10 mL/min	T800–60–150–10 mL/min
		5 mL/min	T800–60–150–05 mL/min

[1] SDRAC: Sorghum Distillery Residue Activated Carbon
[2] Type of SDRAC: see the paper – Lin *et al.* (2015)
[3] T850–60– 90–H_2O: T (Activation temperature) – Activation duration – Flow rate – Activating agent (Steam)
[4] T850–60– 90–030 min: T (Activation temperature) – Activation duration – Flow rate – Time of standing or flow velocity

Water tests

The turbidity was tested by the water turbidity detection method – turbidimetric method, as published by EPA (NIEA W219.52C, 2005a).

The pH value was measured by the inwater hydrogen ion concentration measuring method – electrode method, as published by EPA (NIEA W424.52A, 2008).

The total hardness was tested by the inwater total hardness detection method – EDTA (Ethylenediaminetetraacetic acid) titration, as published by EPA (NIEA W208.51A, 2006). The equation was total hardness (represented by $CaCO_3$, mg/L) = A × B × 1000 / V, where A is the titration volume of EDTA for water specimen titration minus titration volume of Blank; V is the water specimen volume (mL); B is the milligrams of $CaCO_3$ equivalent to 1 mL EDTA volumetric solution.

The nitrite nitrogen was tested by the inwater nitrite nitrogen detection method – spectrophotometry, as published by EPA (NIEA W418.52C, 2013b). The equation is nitrite nitrogen concentration (mg/L) = A × 50 / V, where A is the concentration value (mg/L) measured by calibration curve; V is the water specimen volume (mL). The sodium nitrite standard solution $0.1\,\mu g$/mL was used to discuss the changes in the content adsorbed by SDRAC.

The total bacterial count was tested by the inwater total bacterial count detection method – mixed dilution method, as published by EPA (NIEA E204.55B, 2013a). The equation is total bacterial count (CFU/mL) = total colony counts in the selected culture dish/sum of actual volumes of water specimens in the selected culture dish.

The coliform was tested by the commercial quick test method, namely, Chromocult coliform was used to quickly test the aerobic or facultative anaerobic, gram–negative, and asporulate coliform bacteria. The equation was coliform count (CFU/100mL) = [(red colony + violet colony) × 100] / water specimen volume × extension rate.

The total organic carbon (TOC) was tested by the inwater TOC detection method – peroxy–pyrosulfate thermal oxidation/infrared mensuration, as published by EPA (NIEA W532.52C, 2005a).

Preliminary safety test for water quality

Referring to the Ames Tests, as proposed by Ames *et al.* (1975), Waleh *et al.* (1982), which indicated that if the sample is toxic for the strain, the bacterial count decreases, and the test result is likely to be misjudged. Therefore, the specimen cytotoxicity has to be tested before the mutagenicity test, in order to evaluate whether the growth of the strain is influenced. The mutagenicity can be implemented if there is no toxicity. In this study, the *Salmonella typhimurium* TA98 and TA100 were used as test strains. TA98 is the strain sensitive to frame shifting mutation, as caused by specific mutagens; TA100 is the strain for testing base substitution variation (Mortelmans and Zeiger, 2000). The phosphoric acid buffer solution was used as the blank group (Control), and each water specimen before and after purification by SDRAC was repeated to calculate the colony counts. After cytotoxicity testing, the residual bacterial count has to be higher than 80% of the residual bacterial count of Blank, meaning that the specimen has no toxic response to cells (Waleh *et al.*, 1982). In the mutagenicity test, if the specimen induced by spontaneous revertants is higher than Control by two times, i.e. mutagenicity ratio above 2.0, it means the specimen has mutagenic-

Table 2. Turbidity [1] of water specimen for raw water and slow filter water after processing by standing method with SDRAC

Unit: NTU

Water specimen [2]	Raw water [3]	Percent removal (%) [4]	slow filter water [3]	Percent removal (%)
Blank [5]	20.60 (1.84)[b6)]	–	1.34 (0.39) [b]	–
T850–60–090–10 mL/min	2.50 (0.26) [a]	87.8	1.11 (0.24) [b]	17.1
T850–60–090–05 mL/min	1.73 (0.16) [a]	91.6	0.60 (0.06) [a]	55.2
T800–60–090–10 mL/min	2.80 (0.29) [a]	86.4	1.24 (0.26) [b]	7.4
T800–60–090–05 mL/min	1.96 (0.03) [a]	90.4	0.59 (0.15) [a]	55.9
T800–60–120–10 mL/min	2.36 (0.36) [a]	88.5	0.99 (0.24) [ab]	26.1
T800–60–120–05 mL/min	1.90 (0.14) [a]	90.7	0.61 (0.14) [a]	54.4
T800–60–150–10 mL/min	2.36 (0.28) [a]	88.5	0.94 (0.15) [ab]	29.8
T800–60–150–05 mL/min	1.84 (0.06) [a]	91.0	0.55 (0.04) [a]	58.9

[1] Turbidity of water quality standard for drinking water source is no standard; Turbidity of water quality standard for drinking water is 2 NTU (Environmental Protection Administration, 2013c)

[2] Water specimen: see Table 1

[3] The raw water was obtained on May 27, 2014, and the slow filter water was obtained on May 27, 2014.

[4] Percent removal (%) = [(turbidity of blank – turbidity of water specimen after processing with SDRAC) / turbidity of blank] * 100

[5] Blank: water specimen is raw water or slow filter water that unprocessed with any SDRAC.

[6] Mean (standard deviation) with the different superscripts are significantly different ($\rho < 0.05$) by Duncan's multiple range tests

ity (Ames *et al.*, 1975).

Statistical analysis

The results are represented by mean (standard deviation). The statistical analysis was conducted using SPSS 12 (Statistical Product and Service Solutions) and Duncan's multiple range test ($\rho < 0.05$). Different letters represent significant difference, while the same letter (same subset) represents no significant difference.

RESULTS AND DISCUSSION

Evaluation of water purification
Turbidity

Turbidity is caused by the suspended substances in water, including fine colloidal particles to coarse and dispersed suspended particles (Shi, 2003). Generally, the turbidity of a water specimen is determined by the ratio of the reference standard turbidity suspension of the water specimen to the intensity of specific scattered light, and the unit is Nephelometric Turbidity Unit (NTU). Table 2 showed the results of turbidity of the raw water and slow filter water purified by the filtration method with SDRAC. The raw water turbidity was 20.6 NTU, and the slow filter water was 1.34 NTU. The turbidity of the raw water was reduced to 2.50–1.73 NTU after filtration, the water turbidity was able to be removed by 87.8–91.6%; the turbidity of slow filter water was removed by 7.4–58.9%, i.e. 1.24–0.55 NTU. The turbidity of raw water treated by the standing method was removed by 84.4–87.0%, the turbidity was reduced to 3.02–2.67 NTU; the turbidity of the slow filter water was removed by 35.0–

91.0%, i.e. turbidity was reduced to 0.87–0.12 NTU (results not shown in Table). According to the results of Duncan's multiple range tests, the SDRAC processed results of different standing periods and filtration flow velocities had significant difference. It is indicated that the SDRAC is influenced by them when purifying water, and the water turbidity decreases as the time extends and the flow velocity decreases. The water quality standard for drinking water specifies turbidity as 2 NTU. It is suggested that the raw water specimen is processed by SDRAC at a flow velocity of 5±2 mL/min can reach the water quality standard for drinking water.

pH value

The raw water pH value was subacid 5.86, and the pH value after SDRAC standing increased to 7.76–9.47. The slow filter water pH value was 6.56, and the pH value after activated carbon standing was 8.18–9.69. The pH value also increased after filtration by SDRAC at different flow velocities. The pH value of the filtered raw water specimen was 6.65–8.39, that of the slow filter was 7.04–8.43 (results not shown in Table). The pH value increases because when the activated carbon is heated, the carbon single bonding forms double bonding when the hydrogen atom is removed from the precursor surface. The activated carbon material bottom surface delocalization (π electron) forms a Lewis Bases–like effect, and the activated carbon becomes alkaline (Zhu *et al.*, 2012). When the activation temperature is higher than 800°C, the surface is likely to form multiple basic functional groups, such as the oxygen–containing functional group formed by the oxygen transfer of C–CO$_2$ or C–H$_2$O

Table 3. Total hardness [1] of water specimen for raw water and slow filter water after processing by standing method with SDRAC standing method with SDRAC

Unit: mg/L

Water specimen [2]	Raw water [3]	Percent	slow filter water [3]	Percent removal (%)
Blank [5]	115.5 (4.15) [e3]	–	116.6 (4.71) [g]	–
T850–60–090–030min	63.3 (2.69) [a]	45.1	64.4 (1.56) [b]	44.8
T850–60–090–060min	58.9 (5.61) [a]	49.0	56.7 (2.69) [a]	51.4
T850–60–090–120min	57.8 (4.12) [a]	50.0	55.6 (3.11) [a]	52.3
T800–60–090–030min	84.2 (1.56) [d]	27.0	85.3 (1.56) [f]	26.8
T800–60–090–060min	82.0 (1.56) [cd]	29.0	80.9 (1.56) [ef]	30.6
T800–60–090–120min	79.8 (2.69) [bcd]	30.1	77.6 (3.11) [de]	33.4
T800–60–120–030min	79.5 (5.39) [bcd]	31.2	82.0 (3.11) [ef]	29.7
T800–60–120–060min	78.7 (1.56) [bcd]	31.8	77.6 (1.56) [de]	33.4
T800–60–120–120min	72.1 (1.56) [b]	37.6	73.2 (0.00) [cd]	37.2
T800–60–150–030min	79.8 (2.69) [bcd]	30.9	78.7 (3.11) [de]	32.5
T800–60–150–060min	78.7 (3.11) [bcd]	31.8	76.5 (2.69) [cde]	34.4
T800–60–150–120min	74.3 (4.12) [bc]	35.6	71.0 (1.56) [c]	39.1

[1] Total hardness of water quality standard for drinking water source is no standard; Total hardness of water quality standard for drinking water is 300 mg/L (Environmental Protection Administration, 2013c)

[2], [3], [5], [6] see Table 2

[4] Percent removal (%) = [(total hardness of blank – total hardness of water specimen after processing with SDRAC) / total hardness of blank] * 100

reaction, where the higher activation temperature means the carbon surface contains more basic functional groups (Park and Kim, 2001; Zhu *et al.*, 2012). The pH value specified by the water quality standard for drinking water is 6.0–8.5, the pH value after the treatment by SDRAC in this test is higher than the water standard. However, the pH value after the process by the activated carbon filter tank of the purification plant increases. The subsequent process of the general water treatment plant can make water quality meet the standard (Anu *et al.*, 2006).

Total hardness

The amount of multivalent metal cations of Ca^{2+}, Mg^{2+}, Sr^{2+} and $Fe\ 2^{+}$ in water is the factor determining water hardness, too hard water is inapplicable to industry, and it influences the taste of drinking water. Taiwan's water quality standard for drinking water restricts the total hardness, and the value is 300 mg/L (calculated by $CaCO_3$). Table 3 showed the total hardness of raw water was 115.5 mg/L and that of slow filter water was 116.6 mg/L. The total hardness of raw water from various reservoirs in Kinmen is 58.1–248.5 mg/L (Construction and Planning Agency, 2005). The total hardness of water specimens for this test is in the range. Table 3 also shows the changes in the total hardness of raw water and slow filter water specimens purified by the standing method with SDRAC. The total hardness of raw water was reduced by 27.0–50.0%, the total hardness of T850–60–090–H_2O was reduced to 57.8–63.3 mg/L, the total hardness decreases as the standing time extends, and the slow filter water had the same result. The filtration method also reduced the total hardness to 67.7–88.6 mg/L, the T850–60– 90–H_2O had the maximum decreasing percentage (results not shown in Table). SDRAC can soften water by the standing or filtration method. McCafferty *et al.* (2000) and Lee *et al.* (2003) indicate that the inorganic substance is likely to exchange ions with the carbon surface functional groups, and form the complex for absorption. Therefore, when SDRAC is used for high temperature steam activation, due to the oxygen transfer of C–H_2O reaction and the formation of oxygen–containing and basic functional groups on the carbon surface, SDRAC can reduce the total hardness in water during water treatment. The basic functional groups on the surface increase with the activation temperature, which is helpful to reducing the total hardness of water.

Nitrite nitrogen

Nitrite nitrogen (NO_2–N) is one of the forms of nitrogen dissolved in water. The other forms are ammonia nitrogen (NH_3–N), nitrate nitrogen (NO_3–N), organonitrogen (Org–N), and various forms in the aerobic state (Sawyer, 1967). Nitrite nitrogen is listed as an unhealthy material in the water quality standard for drinking water. It is carcinogenic to the human body, and affects human health and infant methemoglobinemia (Shi, 2003). If it enters human body, it is rapidly reduced to nitrite and generates carcinogenicity. The nitrite nitrogen or nitric nitrogen; therefore, is regarded as a water pollution index, and drinking water quality standards have close restraints

Table 4. Content change of sodium nitrite standard solution 1) during different standing time after processing with SDRAC

Unit: CFU/mL

Water specimen [2]	Raw water [3]	Percent removal (%) [4]
T850–60– 90– 30min	0.065 (0.0)[5]	35
T850–60–90– 60min	0.046 (0.0)	54
T850–60–90–120min	0.021 (0.0)	79
T800–60–90– 30min	0.089 (0.0)	11
T800–60–90– 60min	0.065 (0.0)	35
T800–60–90–120min	0.036 (0.0)	64
T800–60–120– 30min	0.088 (0.0)	12
T800–60–120– 60min	0.060 (0.0)	40
T800–60–120–120min	0.033 (0.0)	67
T800–60–150– 30min	0.064 (0.0)	36
T800–60–150– 60min	0.050 (0.0)	50
T800–60–150–120min	0.029 (0.0)	71

[1] Sodium nitrite standard solution: 0.1 µg/mL
[2] see Table 1
[3], [5] see Table 2
[4] Percent removal (%) = [(sodium nitrite standard solution (0.1 µg/mL) – sodium nitrite standard solution) / sodium nitrite standard solution (0.1 µg/mL)] * 100

for nitrite nitrogen and nitric nitrogen (EPA, 2013). The nitrite nitrogen concentration in the raw water and slow filter water of this study was 0.01 mg/L. The slow filter water was 0.00 mg/L, which was lower than 0.1 mg/L of the water quality standard for drinking water, thus, nitrite nitrogen was not tested. Therefore, this study used sodium nitrite standard solution 0.1 µg/mL in the standing method to discuss the changes in the content adsorbed by SDRAC. The results are as shown in Table 4. The sodium nitrite standard solution 0.1 µg/mL after standing filtration was reduced to 0.089–0.021 µg/mL, and the percent removal was 11.0–79.0%. It is said that SDRAC can adsorb nitrite nitrogen, which is possibly due to the basic (alkaline) functional group (Cabal *et al.*, 2009; Nunell *et al.*, 2012).

Total bacterial count

Table 5 showed that the total bacterial count of raw water and slow filter water specimens, as filtered by the standing method with SDRAC. The total bacterial count of raw water was 1539 CFU/mL, the total bacterial count decreased significantly after standing, and the total bacterial count in water was able to be reduced by 99.8–99.9% after 30 min SDRAC standing. The total bacterial count was 1–3 CFU/mL, and the total bacterial count increased with the standing time. The total bacterial count after 120 min SDRAC standing was 10–16 CFU/mL; that of slow filter water was 1025 CFU/mL, and the value was 1–28 CFU/mL after standing filtration. The total bacterial count was able to be reduced by 97.2–99.9%, thus, conforming to the water quality standard for drink-

Table 5. Total bacterial count [1] of water specimen for raw water and slow filter water after processing by standing method with SDRAC

Unit: mg/L

Water specimen [2]	Raw water [3]	Percent	slow filter water [3]	Percent removal (%)
Blank [5]	1539 (66.00) [b6]	–	1025 (104.00) [b]	–
T850–60–090–030min	3 (0.82) [a]	99.8	1 (0.00) [a]	99.9
T850–60–090–060min	4 (1.70) [a]	99.7	5 (1.63) [a]	99.5
T850–60–090–120min	16 (3.40) [a]	98.9	25 (2.05) [a]	97.6
T800–60–090–030min	1 (0.00) [a]	99.9	1 (0.47) [a]	99.9
T800–60–090–060min	4 (2.16) [a]	99.7	2 (0.82) [a]	99.8
T800–60–090–120min	10 (0.47) [a]	99.3	12 (2.49) [a]	98.8
T800–60–120–030min	2 (0.47) [a]	99.8	1 (0.47) [a]	99.9
T800–60–120–060min	4 (1.63) [a]	99.7	5 (1.63) [a]	99.5
T800–60–120–120min	15 (2.62) [a]	99.0	28 (3.40) [a]	97.2
T800–60–150–030min	1 (0.00) [a]	99.9	1 (0.47) [a]	99.9
T800–60–150–060min	2 (0.88) [a]	99.8	3 (2.16) [a]	99.7
T800–60–150–120min	13 (2.05) [a]	99.1	20 (2.01) [a]	98.0

[1] Total bacterial count of water quality standard for drinking water source is no standard; Total bacterial count of water quality standard for drinking water is 100 mg/L (Environmental Protection Administration, 2013c)

[2], [3], [5], [6] see Table 2

[4] Percent removal (%) = [(total bacterial count of blank – total bacterial count of water specimen after processing with SDRAC) / total bacterial count of blank] * 100

ing water (100 CFU/mL). Ogawa *et al.* (2011) indicates that during standing in activated carbon, the total bacterial count in water varies with the activated carbon measure. The total bacterial count in water effectively decreases as the activated carbon measure is increased. Uraki *et al.* (2008) reported that the activated carbon filtration and standing process are effective methods to reduce the total bacterial count in water. The active mechanism is that the strongly alkaline solution condition is presented in activated carbon to cause bacteria death and the bacteria are adsorbed in the activated carbon.

In terms of filtration method (results not shown in Table), the total bacterial count in the filtered raw water was 381–436 CFU/mL; that in the slow filter water was 248–333 CFU/mL, and the total bacterial count increased as the filtration flow velocity decreased. According to the above results, the standing or filtration test of SDRAC showed that the total bacterial count in water increased slightly with the SDRAC contact time (standing time or filtration flow velocity). Because the activated carbon pores are adsorptive and developed, it is advantageous to the adhesion and growth of microorganisms, and the colonies increase in the activated carbon with the filtration or standing time (Liao *et al.*, 2013). The increase of colonies with the activated carbon contact time is a common phenomenon in the activated carbon filter of a purification plant. General purification plants often set the sterilization procedure behind the activated carbon filter in order to increase sterilization efficiency. Many studies have reported that the Alphaproteobacteria extensively exists in the activated carbon filter for drinking water

treatment (Ko *et al.*, 2007; Magic–Knezev *et al.*, 2009; Liao *et al.*, 2012). Thus, in the water purification procedure of the purification plant, the activated carbon filter is mostly followed by the ozone sterilization procedure in order to control the quantity of aquatic microorganisms (Chen and Chang, 2005).

To sum up, in the standing or filtration treatment of SDRAC, its alkaline characteristics can effectively reduce the total bacterial count in water, but as the contact time extends, the developed pore structure is contrarily helpful to the adhesion and growth of microorganisms.

Coliform

Coliform often exists in human and homeothormous excreta, and is highly correlated with feces source pollution. It is a pollution index bacterium, and it is the fecal pollution level of drinking water. Thus, it is specified in the water quality standard for drinking water of Taiwan. It is 20 CFU/mL in the water quality standard for drinking water sources, and 0.06 CFU/mL in the water quality standard for drinking water (EPA, 1997; 2013c). The coliform in raw water and slow filter water for this test is 3 and 22 CFU/mL, respectively, meaning that the raw water of the Jinsha reservoir and the slow filter water of the Ronghu water process plant have no high fecal pollution, and conform to the water quality standards for drinking water sources and drinking water. The filtrate of raw water and slow filter water filtered by SDRAC and standing method or filtration method was below 1 CFU/mL (results not shown in Table). Therefore, SDRAC at a high activation temperature has better results in the water purification process for subsequent testing. The T850–

Table 6. Total organic carbon [1] of water specimen for raw water and slow filter water after processing by standing method with 2 types of SDRAC

Unit: mg/L

Water specimen [2]	Raw water [3]	Percent	slow filter water [3]	Percent removal (%)
Blank [5]	7.6	–	8.7	–
T850–60–90–30min	5.1	32.8	5.7	34.5
T850–60–90–10mL/min	5.6	26.3	6.6	24.1

[1] Total organic carbon (TOC) of water quality standard for drinking water source is no standard; TOC of water quality standard for drinking water is 100 mg/L (Environmental Protection Administration, 2013c)

[2), 3), 5), 6)] see Table 2

[4] Percent removal (%) = [(TOC of blank – TOC of water specimen after processing with SDRAC) / TOC of blank] * 100

60–90–H$_2$O at an activation temperature of 850°C is used to assess the TOC in water and water safety.

TOC

The organic matter in water has adverse effect on the colority, smell, and taste of drinking water, and it influences the dosage of coagulant in the water purification procedure, interfering in advanced water process and heavy metals Fe and Mn in water form complex, thus, increasing the content of heavy metals, corrosion of distribution lines, and regrowth of microorganisms in water. In the chlorination procedure, it is likely to react with disinfector to produce the sterilization byproducts of haloform and halothane, affecting human health (Bull, 1982; Jacangelo *et al.*, 1995). Therefore, the content of TOC is considered in the water purification procedure, and TOC in the standard of drinking water sources is 4 mg/L. Table 6 showed the changes in the TOC in water after filtration and standing of raw water and slow filter water by T850–60–090–H$_2$O. The TOC of raw water was 7.6 mg/L, after 30 min standing, the TOC removal rate was 32.8%, reduced to 5.1 mg/L. The percent removal of the filtration method was 26.3%, and the value decreased to 5.6 mg/L. There is adsorption effect on the TOC of slow filter water. The TOC of slow filter water was 8.7 mg/L.

After SDRAC standing, the TOC in water was 5.7 mg/L; that of filtration method was 6.6 mg/L, the TOC percent removal was 24.1–34.5%. Anu *et al*. (2006) studied the bioactive carbon filter of the Finland purification plant, and evaluated the activated carbon filter treatment by different methods or conditions. They found that the natural organic matter in the raw water was removed by 53–95%, and the water was purified. It is suggested that the effect of SDRAC on removing or reducing TOC is related to the standing time and filtration flow velocity.

Safety assessment of water quality
Cytotoxicity

The cytotoxicity of water specimens after 30 min T850–60– 90–H$_2$O standing and filtration method at the flow velocity of 10 mL/min for *Salmonella typhimurium* TA98 and TA100 strains was tested. The results are as shown in Table 7. The bacterial count of Control without rat liver enzyme mixture (S9 mix) was 1663 in TA98, 1979 in TA100; the TA98 with S9 mix was 1836, TA100 was 2140. The bacterial count of raw water without the S9 mix was 1715 in TA98, and 1983 in TA100. Survival was 100–103%. The TA98 with the S9 mix was 1875, and TA100 was 2250. Survival was 102–105%. Survival of slow filter water without the S9 mix was 101–104% in

Table 7. Cytotoxicity of water specimen for raw water and slow filter water after processing by standing and filtration methods with 2 types of SDRAC toward *Salmonella typhimurium* TA98, TA100 with or without S9 mixture

Water specimen [1]		without S9 mixture				with S9 mixture			
		TA98	Survival [2] (%)	TA100	Survival (%)	TA98	Survival (%)	TA100	Survival (%)
	Control [3]	1663 (44)[5]	100	1979 (48)	100	1836 (82)	100	2140 (55)	100
raw water	Blank [4]	1715 (45)	103	1983 (26)	100	1875 (53)	102	2250 (90)	105
	T850–60–90–30 min	1741 (29)	104	2030 (32)	102	1882 (54)	102	2328 (66)	108
	T850–60–90–10 mL/min	1844 (50)	110	2036 (33)	102	1963 (69)	106	2340 (54)	109
slow filter water	Blank	1738 (41)	104	2010 (16)	101	1878 (59)	102	2192 (11)	102
	T850–60–90–30 min	1746 (41)	104	2031 (33)	102	1891 (36)	102	2233 (59)	104
	T850–60–90–10 mL/min	1799 (56)	108	2004 (37)	101	1872 (30)	101	2254 (37)	105

[1] see Table 1

[2] Survival (%) = [(the bacterial count of test group / the bacterial count of control group)] * 100

[3] Control: only with 0.1 mL phosphate buffer saline

[4] see Table 2

[5] Mean (standard deviation)

Table 8. Mutagenicity of water specimen for raw water and slow filter water after processing by standing and filtration methods with 2 types of SDRAC toward *Salmonella typhimurium* TA98, TA100 with or without S9 mixture

Water specimen [1]		without S9 mixture				with S9 mixture			
		TA98	MR[2]	TA100	MR	TA98	MR	TA100	MR
Control [3]		52 (4)[5]	1.00	145 (4)	1.00	53 (7)	1.00	165 (10)	1.00
raw water	Blank [4]	51 (7)	0.98	141 (9)	0.97	53 (6)	1.00	167 (9)	1.01
	T850–60–90–30 min	50 (8)	0.96	147 (9)	1.01	59 (8)	1.11	168 (6)	1.02
	T850–60–90–10 mL/min	53 (5)	1.02	160 (4)	1.10	55 (3)	1.03	157 (5)	0.95
slow filter water	Blank	51 (8)	0.98	148 (5)	1.02	53 (7)	1.00	164 (5)	0.99
	T850–60–90–30 min	46 (5)	0.88	128 (6)	0.88	52 (4)	0.98	163 (5)	0.98
	T850–60–90–10 mL/min	48 (6)	0.86	146 (6)	1.00	52 (7)	0.98	165 (11)	1.00

[1] see Table 1
[2] MR (Mutagenicity ratio) = induced revertants per plate/spontaneous revertants per plate (Control)
[3] Control: see Table 7
[4] see Table 2
[5] Mean (standard deviation)

TA98, while that with S9 mix was 102%. Therefore, the water specimens have no cytotoxicity. In order to guarantee the safety of water specimens processed by SDRAC, the cytotoxicity of the water specimens processed by SDRAC was tested. The same Table showed that after 30 min standing and filtration at flow velocity of 10 mL/min, the raw water processed by T850–60– 90–H_2O was 1741–1844 in TA98 without the S9 mix, and Survival was 104–110%; that in TA100 was 2032–2036, Survival was 102%; that with the S9 mix was 1882–1963 in TA98, Survival was 102–106%; TA100 was 2328–2340, Survival was 108–109%. Survival of slow filter in TA98 and TA100, with or without the S9 mix, was 101–108%. It is said that that Survival of the processed water specimens in TA98 and TA100 strain tests is higher than 80% of Control, there is no cytotoxicity, and the mutagenicity can be carried out.

Mutagenicity

The results of mutagenicity are as shown in Table 8. The revertants of *Salmonella typhimurium* TA98 and TA100 induced by raw water and slow filter water, with and without the S9 mix, had not exceeded two times the spontaneous revertants of Control, meaning the raw water and slow filter water had no mutagenicity. The same Table also showed that, in the mutagenicity results of water specimens processed by SDRAC, the water specimens of raw water and slow filter water after 30 min standing and filtration at flow velocity of 10 mL/min in T850–60– 90–H_2O had no mutagenicity, and did not exceed two times the spontaneous revertants of Control. It is said that that the raw and slow filter water before and after purification have no mutagenicity toward *S. typhimurium* TA98 and TA100.

CONCLUSIONS

This study evaluated the feasibility of SDRAC for purifying water, and performed the Ames Tests to ensure the preliminary safety of water quality before and after purification. In terms of water turbidity testing, the SDRAC prepared in different conditions could reduce water turbidity by 7.4–91.6%. The value was 0.12–2.93 NTU. The pH value in water was increased by SDRAC filtration or standing in solution, and the pH value after processing was increased by 13.4–61.6 %. The total hardness of the raw water and slow filter water specimens processed by SDRAC was removed by 23.2–52.3%, and the value was 55.6–88.6 mg/L. The nitrite nitrogen concentration could be reduced after processing by SDRAC. After the SDRAC process, the total bacterial count in water was removed by 67.5–99.9%, and the percent removal of coliform was 99.9%. The water tests conform to the water quality standard for drinking water in Taiwan. SDRAC could remove 26.3–34.5% of TOC. The bacterial survival rate of cytotoxicity testing for water specimens before and after purification was higher than 80% of Control, meaning there was no cytotoxicity. The mutagenicity results showed that the spontaneous revertants were not exceeded by two times, thus, there were no mutagenicity. The water processed by SDRAC can meet the water quality standard for drinking water, and the preliminary safety of SDRAC can be guaranteed by the Ames Tests. Therefore, SDR may be used to prepare activated carbon, which can be prepared into SDRAC, and is more functional to purify water.

REFERENCES

Ames, B. N., J. McCann and E. Yamasaki 1975 Methods for detecting carcinogens and mutagens with the *salmonella*/mammalian microsome mutagenicity test. *Mutation research*, **31**: 347–364

A Andersson, A. and A. Kehn 2001 Impact of temperature on nitrification in biological activated carbon filters used for drinking water treatment. *Water research*, **35**(12): 2923–2394

Anu, M., N. Vieno and T. Tuhkanen 2006 Efficiency of the activated carbon filtration in the natural organic matter removal. *Environment International*, **32**(3): 324–331

Bull, R. J. 1982 Health effect of drinking water disinfectants and disinfection by products. *Science andTtechnology*, **16**(10): 554–561

Cabal B, T. Budinova, C. O. Ania, B. Tsyntsarski, J. B. Parra and B. Petrova 2009 Adsorption of naphthalene from aqueous solution on activated carbons obtained from bean pods. *Journal of Hazardous Materials*, **161**(2–3): 1150–1156

Chen, C. H. and C. H. Change 2005 Tap Water (in Chinese). Sintech Foundation for Research and Development of Engineering Science and Technologies, pp. 6–66

Construction and Planning Agency 2005 *Long–term Environmental Monitoring in Kinmen National Park (IV)* (in Chinese). Construction and Planning Agency (CPA), Ministry of the Interior, Taiwan ROC

Environmental Protection Administration 1997 *Water Quality Standard for Drinking Water Source*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Environmental Protection Administration 2005a *NIEA W219.52C*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Environmental Protection Administration 2005b *NIEA W532.52C*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Environmental Protection Administration 2006 *NIEA W208.51A*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Environmental Protection Administration 2008 *NIEA W424.52A*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Environmental Protection Administration 2013a *NIEA E204.55B*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Environmental Protection Administration 2013b *NIEA W418.52C*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Environmental Protection Administration 2013c *Water Quality Standard for Drinking Water*. Environmental Protection Administration (EPA), Executive Yuan, Taiwan ROC

Jacangelo, J. G., J. DeMarco, D. M. Owen and S. J. Randtke 1995 Selected process for removing NOM: overview. *American Water Works Association*, **87**(1): 64–77

Jiang, C. T. 2010 *Application Principle and Technology of Activated Carbon*. Chemical Industry Publishing, pp. 5–10

Kihn, A., P. Laurent and P. Servais 2000 Measurement of potential activity of fixed nitrifying bacteria in biological filters used in drinking water production. *Journal of industrial microbiology and biotechnology*, **24**(3): 161–166

Ko, Y. S, Y. J. Lee and S. H. Nam 2007 Evaluation of a pilot scale dual media biological activated carbon process for drinking water. *Korean Journal of Chemical Engineering*, **24**(2): 53–60

Kunio, E., F. Li, Y. Azuma, A. Yuasa and T. Hagishita 2001 Pore distribution effect of activated carbon in adsorbing organic micropollutants from natural water. *Water Research*, **35**(1): 167–179

Lee, S. H., W. Nishijima, C. H. Lee and M. Okada 2003 Calcium accumulation on activated carbon deteriorates synthetic organic chemicals adsorption. *Water Research*, **37**(19): 4631–4636

Liao, X. B., C. Chen, C. H. Chang, Z. Wang, X. J. Zhang and S. G. Xie 2012 Heterogeneity of microbial community structures inside the up–flow biological activated carbon (BAC) filters for the treatment of drinking water. *Biotechnology and Bioprocess Engineering*, **17**(4): 881–886

Liao, X., C. Chen, Z. Wang, R. Wam, C. H. Chang, X. J. Zhang and S. G. Xie 2013 Changes of biomass and bacterial communities in biological activated carbon filters for drinking water treatment. *Process Biochemistry*, **48**(2): 312–316

Lin, H. C., J. S. Hu, W. J. Lee, C. W. Peng, Y. J. Lai, S. C. Wu and N. Fujimoto 2015 Adsorption characteristics and pore structure of activated carbons prepared from sorghum distillery residue. *Journal of the Faculty of Agriculture Kyushu University*, Japan. (*In pressed*)

Magic–Knezev, A., B. Wullings and K. D. Vander 2009 Polaromonas and hydrogenophaga species are the predominant bacteria cultured from granular activated carbon filters in water treatment. *Journal of Applied Microbiology*, **107**(5): 1457–1467

Marguerite, M. B., M. C. Hascoet, Y. Levi, F. Erb, and N. Pommeryt 1986 Role of ozone and granular activated carbon in the removal of mutagenic compounds. *Environmental Health Perspectives*, **69**: 159

Maron, D. M. and B. N. Maron 1983 Revised methods for the *Salmonella* mutagenicity Test. *Mutation Research*, **113**: 173–215

McCafferty, N. D., M. E. Callow, L. Hoggert, B. Holden and B. S. C. Leadbeater 2000 Application of method to quantify carbonate precipitated on granular activated carbon used in potable water treatment. *Water Research*, **34**(8): 2199–2206

Monarca, S., J. R. Meier and R. J. Bull 1983 Removal of mutagens from drinking water by granular activated carbon–Evaluation using bacterial mutagenicity tests. *Water Research*, **17**(9): 1015–1026

Mortelmans, K. and E. Zeiger. 2000 The Ames *Salmonella* / microsome mutagenicity assay. *Mutation Research*, **455**(1): 29–60

Nunell, G. V., M. E. Fernández, P. R. Bonelli and A. L. Cukierman 2012 Conversion of biomass from an invasive species into activated carbons for removal of nitrate from wastewater. *Biomass and Bioenergy*, **44**: 87–95

Ogawa, M., T. B. Bardant, Y. Sasaki, Y. Tamai, S. Tokura and Y. Uraki 2011 Electricity–free production of activated carbon from biomass in borneo to improve water quality. *Bio resources*, **7**(1): 236–245

Omri, A., M. Benzina and N. Ammar 2013 Preparation, modification and industrial application of activated carbon from almond shell. *Journal of Industrial and Engineering Chemistry*, **19**(6): 2092–2099

Park, S. J. and C. K. D. Kim 2001 Influence of activation temperature on absorption characteristic of activated carbons fiber composites. *Carbon*, **39**:1741–1746

Przepiorski, J. 2006 Enhanced adsorption of phenol from water by ammonia–treated activated carbon. *Hazardous Materials*, **135**(1): 453–456

Sawyer, C. N. 1967 *Chemistry for Sanitary Engineers*. New York

Seyed A. D., T. Karanfil, and W. Cheng 2004 Tailoring activated carbons for enhanced removal of natural organic matter from natural waters. *Carbon*, **42**(3): 547–557

Shi, F. C. 2003 *Water Quality Analysis and Detection* (in Chinese). Wun Ching Developmental Publishing, pp. 87–485

Shiu, S. K. 2000 *Introduction to Water Source and Sustainable Development* (in Chinese). Water Resources Agency, Ministry of Economic Affairs, Taiwan ROC, pp. 132–137

Tomaszewska, M. and S. Mozia 2002 Removal of organic matter from water by PAC/UF system. *Water Research*, **36**(16): 4137–4143

Uraki,Y., Y. Tamai, M. Ogawa, S. Gaman and S. Tokura 2008 Preparation of activated carbon from peat. *Bio Resources*, **4**(1): 205–213

Villacan F., M. F. Pereira, and J. M. Jose 2006 Adsorption of simple aromatic compounds on activated carbon. *Colloid and Interface Science*, **293**(1): 128–136

Waleh, N. S., S. J. Rapport and K. E. Mortelmans 1982 Development of a toxicity test to be coupled to the ames salmonella assay and the method of constriction of the required strains. *Mutation Research*, **97**(4): 247–256

Zhou, K. C. and K. T. Lu 2010 Improvement of drinking water qualities by using moso bamboo activated carbon made from different activation temperatures (in Chinese). *Quart. Journal Forest Research of Taiwan*, **32**(4): 35–44

Zhu, Y., J. Gao, Y. Li, F. Sun J. Gao S. Wu and Y. Qin 2012 Preparation of activated carbons for SO_2 adsorption by CO_2 and steam activation. *Journal of Taiwan Institute of Chemical Engineers*, **43**(1): 112–118