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# Assessment of cockle shells, walnut shells, and gingko shells as filter media for low impact development technologies

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## ABSTRACT

In this study, biowaste such as walnut shells (WS), cockle shells (CS), and gingko shells (GS) was evaluated for applicability as soil filter media in low impact development (LID) techniques. Biowastes used in the experiment showed light-weight, high porosity, and high permeability characteristics compared with sand or gravel which are conventionally used in LID. Especially, CS and WS showed high non-point pollutant removal efficiency due to protrusion of filter media surface and macro- and micro-porous. CS were found to be suitable for the infiltration capacity, as it showed high filtration and pollutant reduction. WS exhibited high heavy metal removal performance but was found to have possibility of organic matter leaching. GS were evaluated to be adaptable to green roof as filter media with light-weight and constant filtration capability. The findings of this research are useful for the application of biowaste as filter media in LID techniques.

*Keywords:* Biowaste; Cockle shells; Gingko shells; LID filter media; Walnut shells

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## 1. Introduction

Human activities such as urbanization increased the discharge of various non-point source pollutants such as particulate matters, organic matters and heavy metals. During dry days, non-point source pollutants are accumulated on impervious surfaces and discharged to water bodies along with the stormwater runoff [1,2]. Generally, low impact development (LID) techniques were applied to solve hydrological and environmental problems such as water cycle disruption, water pollution, and soil pollution due to urbanization [3]. LID focuses on the water cycle restoration and minimizes the environmental problems brought about by urban development. The mechanisms of pollutant removal and volume reduction in LID include retention, infiltration, evapo-transpiration, filtration, adsorption and biodegradation [4]. These mechanisms are performed in LID techniques through components such as plants, microorganisms, soil and filter media. In LID techniques, particulate matters were reduced via physical treatment such as filtration, adsorption, and sedimentation. Nutrients such as total nitrogen and total phosphorus are reduced through degradation by microorganism, nitrification and denitrification processes and uptake through plant roots. Heavy metals, on the other hand, were reduced through physico-chemical and biological treatment via soil, filter media and plants [5]. Filter media, being one of the main components of a LID technology, restores the natural water cycle through functions including retention and infiltration during storm events. The range of sizes and filter media surfaces was also found to have affected the physico-chemical mechanisms to reduce pollutants and to act as habitat for microorganisms. The identification of the optimum filter media size ranges and surface area is important to improve the physical, chemical and biological mechanisms through filter media. Filter media applied to the LID technique can be classified into inorganic filter media such as gravel, sand, zeolite, bottom ash and organic filter media such as woodchip, bark and other mulching materials. In general, inorganic filter media were applied

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to the bottom layer of the LID facility to detain and retain more waters for infiltration. These inorganic filter media have high removal efficiency due to its adsorption and filtration capabilities and play an important role in the removal of pollutants. However, use of such filter media in LID technologies could impose higher cost implication. Generally, inorganic filter media applied to LID were heavy indicating that high maintenance costs were required when replacing these filter media. In addition, the sand and gravel media, which are widely applied to LID facilities, have very low biological function and little pollutant removal efficiency due to the absence of micro-pores. On the other hand, organic filter media have high efficiency in pollutant removal due to their filtration and adsorption functions, but there could be some settlement problem due to biodegradation of organic materials. Various filter media researches were being conducted to overcome the disadvantages of the filter media. For example, oyster shells produce about 280,000 tons of marine waste per year, but less than 10% of them are processed for fertilizer and 80%–90% are landfilled without being recycled. The neglected oyster shells caused environmental problems such as pollution of coastal areas, difficulty in managing public wastes and damage to natural landscapes [6]. Research on biowaste use has been carried out in the fields of construction materials, environmental industrial recycling and recycling as food. In developed countries, biosorption of organic filter media have been studied [7,8]. Wastes produced from fishery industries such as oyster shells, and mollusk shells were extensively used as adsorbent and thus showed high performance for the removal of organic and inorganic pollutants [9–12].

The filtration capability of the oyster shells, an example of biowaste, was found in various researches and generally shows an average removal efficiency of 85%–89% for SS and 80%–85.1% for COD [13–15]. Assessment of biowastes applicability for wastewater treatment has been applied in various studies, but studies on applicability of these biowaste as filter media to non-point source pollutant management during storm events were insufficient. Therefore, this study evaluated the applicability of biowastes light-weight materials including cockle shells (CS), walnut shells (WS) and gingko shells (GS) as filter media of LID facilities.

## 2. Materials and methods

### 2.1. Physical and chemical characteristics of biowaste

Fig. 1 shows the research and experimental scenarios, monitoring and analyses conducted in the biowaste. Each biowaste filter media with size ranging from 0.5 to 1 cm and 1 to 2 cm were evaluated considering the appropriate porosity of the LID facility of 0.35 or more. Each media was cleaned by washing three times or more to remove foreign substance from the filter media surface area. The filter media were dried naturally for more than 3 d. The particle sizes were classified using the standard sieving test using standard after drying, then was analyzed for unit weight, porosity and coefficient of permeability. Desorption experiment was performed to evaluate the decay and desorption capability of biowaste. During the desorption experiment, 50 g of biowastes were placed in 500 mL of distilled water and stored in a JSBI-250C maintained at 20°C for a total of 48 h.

### 2.2. Column test and data analysis

Column testing, as shown in Fig. 2, was performed to evaluate the applicability of biowaste as filter media in LID technologies. The acrylic column was made with 10 cm diameter, 100 cm height, and 0.7 cm diameter holes at the bottom of column. The biowaste filter media was filled up to 23 cm from the bottom of the column and the inflow rate was 0.002 $cm^3/s$, which were determined as based on LID design guidelines of Korean Ministry of Environment. The guidelines are required to determine the LID sizing with 80% of the cumulative rainfall during 10 y in the watershed [16]. The remaining 77 cm of the column was utilized as ponding depth in cases of low infiltration. The hydraulic operating conditions of each column are summarized in Table 1. The synthetic stormwater runoff used in each test run was prepared by diluting road sediment of 150 μm or less with tap water at 1:1 and used synthetic stormwater runoff resulting to a total suspended

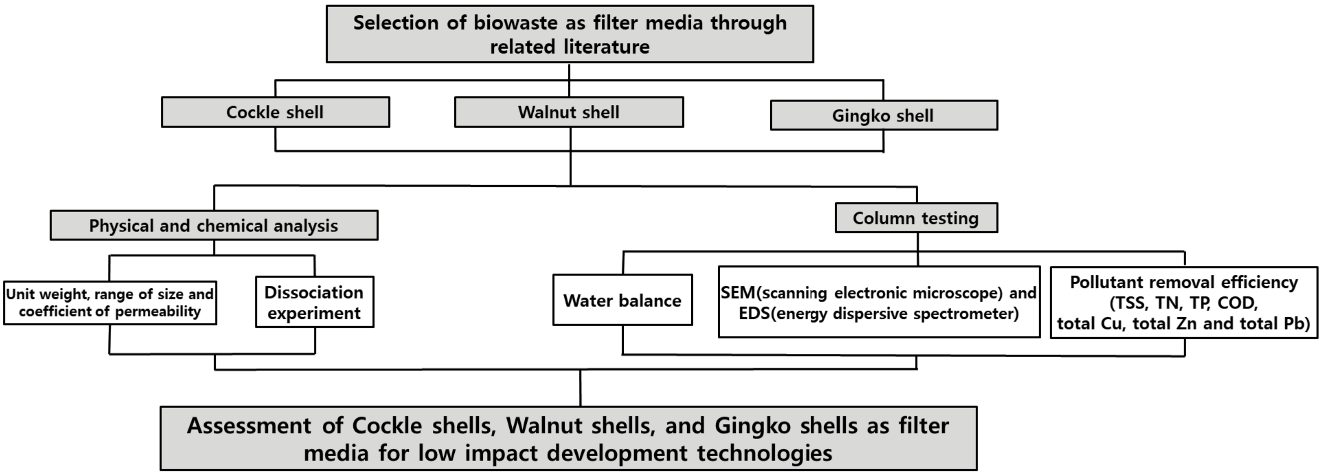

Fig. 1. Research and experimental scenarios, monitoring and analyses.

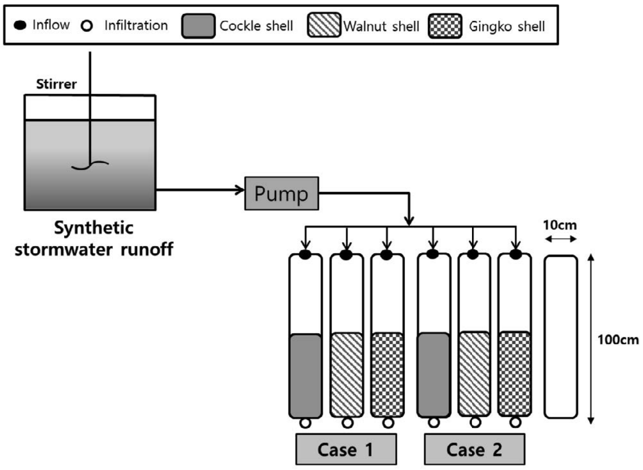

Fig. 2. Schematic design of column for testing.

Table 1
Monitoring condition for each column

| Property | Case 1 | Case 2 |
|---|---|---|
| Filter media size, cm | 0.5–1 | 1–2 |
| Inflow rate, $cm^3/s$ | 0.02 | |
| ADD, d | 3 | |
| Operating time, h | 2 | |
| Flow check time, min | Inflow: every 10<br>Outflow: every 5 | |
| Sampling time, min | 0, 30, 60 and 120 | |
| Water quality analysis | TSS, COD, TN, TP and heavy metal | |

solid (TSS) concentration of 110 mg/L (Table 2). The inflow rate was measured every 10 min to minimize the errors that occurred during the measurement. The infiltration rate was measured every 5 min as soon as infiltration occurred at the sampling port. Water samples were analyzed for water constituents including TSS, total nitrogen (TN), total phosphorus (TP), chemical oxygen demand (COD) and total heavy metals including copper (total Cu), cadmium (total Cd) and lead (total Pb) according to the standard method for the examination of water and wastewater [17]. Scanning electronic microscope (SEM) analysis and energy dispersive spectrometer (EDS) analyses were performed for comparison and analysis of surface characteristics before and after the column tests.

Eq. (1) shows the water balance for the assessment of water cycle [18]. $\text{Vol}_{\text{run}}$ represents the runoff volume; $\text{Vol}_{\text{dis}}$ represents the influent volume; $\text{Vol}_{\text{infil}}$ represents the infiltrated volume; $\text{Vol}_{\text{ret}}$ represents the retained volume; $\text{Vol}_{\text{sto}}$ represents the accumulated volume in the column and $\text{Vol}_{\text{loss}}$ represents the other lost volume.

$$\text{Vol}_{\text{run}} - \text{Vol}_{\text{dis}} = \text{Vol}_{\text{infil}} + \text{Vol}_{\text{ret}} + \text{Vol}_{\text{sto}} + \text{Vol}_{\text{loss}} \quad (1)$$

The average concentration of storm water runoff was calculated by using the event-weighted concentrations (EMC) and removal efficiency as shown in Eqs. (2) and (3), respectively. $C(t)$ and $Q_{\text{TR}_u}(t)$ are the pollutant concentrations and outflow rates for the duration of storm water runoff.

Table 2
Synthetic stormwater influent concentrations (mean ± standard deviation, mg/L)

| TSS | 223 ± 23 |
|---|---|
| COD | 100 ± 12 |
| TN | 2 ± 2 |
| TP | 1 ± 1 |
| Total Cu | 0.34 ± 0.022 |
| Total Zn | 0.14 ± 0.1 |
| Total Pb | 0.34 ± 0.01 |

$$\text{EMC}\left(\text{mg/L}\right) = \frac{\int_0^T C(t) \times Q_{\text{TR}_u}(t)\,dt}{\int_0^T Q_{\text{TR}_u}(t)\,dt} \quad (2)$$

$$\text{Removal efficiency} = \frac{\text{Average influent EMC} - \text{Average effluent EMC}}{\text{Average influent EMC}} \quad (3)$$

## 3. Results and discussion

### 3.1. Analysis of physical and chemical characteristic of biowastes

#### 3.1.1. Physical characteristic analysis

Table 3 are the physico-chemical characteristics of biowaste such as CS, WS and GS. The CS was found to have the highest average unit weight of 780 ± 62 $kg/m^3$ among the biowaste, followed by WS and GS. CS was found to have about three times higher unit weight than other media, but about 60% lower than gravel. The average porosity of GS was 0.48 while the coefficient of permeability ranging from 24 to 26 m/h was similar to WS. Comparing the permeability coefficients of typical LID filter media and biowaste filter media gravel and woodchip were found to have higher permeability coefficient compared with biowaste filter media whereas sand was found to have lower permeability coefficient. Considering porosity, woodchip media were found to be highest at 0.75 while sand was the found to have lowest porosity equal to 0.31. In the case of biowaste, the GS was found to have highest porosity of 0.48 which was 35% lower than the porosity of woodchip. CS, being an inorganic filter media, was found to have porosity of 0.4 which was 25% and 20% lower than the porosity of gravel and sand, respectively. The particle sizes of the biowaste media affect the unit weight, porosity and coefficient of permeability of the media. Smaller the particle sizes implied higher unit weight, and lower porosity and permeability coefficient. The porosity and coefficient of permeability of biowaste were larger than sand and was found to be applicable to LID facilities with high functions water cycle. In addition, the unit weight of biowaste was evaluated to be more than 55%–60% lower than that of gravel. This result showed that maintenance costs could be reduced when applying biowaste as filter media in LID facilities.

Table 3
Physical characteristics of natural filter media

| | Cockle shell (CS) | | Walnut shell (WS) | | Gingko shell (GS) | | Sand [19] | Gravel [19] | Woodchip, (crushed) |
|---|---|---|---|---|---|---|---|---|---|
| | Case 1 | Case 2 | Case 1 | Case 2 | Case 1 | Case 2 | | | |
| Range of size, cm | 0.5–1 | 1–2 | 0.5–1 | 1–2 | 0.5–1 | 1–2 | 0.3–2.4 | 4.8–13.2 | 2.0–4.8 |
| Unit weight, ton/m$^3$ | 0.84 | 0.72 | 0.33 | 0.28 | 0.29 | 0.26 | 1.7–1.8 | 2.0 | 0.43 |
| Porosity | 0.37 | 0.42 | 0.34 | 0.40 | 0.47 | 0.49 | 0.31 | 0.54 | 0.75 |
| Permeability, m/h | 25.42 | 27.29 | 18.83 | 27.94 | 20.59 | 30.71 | 15.66 | 46.51 | 36.54 |

### *3.1.2. Desorption experiment of filter media*

Organic filter media such as WS and GS can affect the nutrient and organic matter concentration during desorption because of the existence of membrane in the filter media. Fig. 3 showed the results of the desorption experiment performed to analyse the effects of the media on the pollutant discharge concentration.

During the experiment, the concentration of COD, TN and TP increased rapidly due to the release of organic materials within the initial 12 h, but the concentration decreased after 24 h. The average COD amounting to 0.17 mg/m$^2$-d was found in WS while it was observed to be lowest in CS amounting to 0.04 mg/m$^2$-d. The COD generated during the initial 12 h was about 0.26, 0.06 and 0.23 mg/g for WS, CS and GS, respectively. However, after 48 h, the COD in WS and GS was found to have similar value of 0.32 mg/g, while CS was found to have the lowest value. During the first 12 h, release of COD and TN increased due to the removal of membrane while it was analyzed that after 12 h the concentration of TN and TP no longer increased due to saturation and full desorption of pollutants. Among the three filter media, GS showed high amounts of COD and TN due to the easy desorption of the membrane inside.

On the other hand, CS had the lowest adsorption of pollutants among the three biowastes because of its mineral compositions. Based on these results, cleaning of biowaste is necessary prior to application in the LID facilities. The decomposition caused by the long-term use of organic filter media was projected to be able to provide carbon sources and nutrients needed for microbial growth of LID facilities.

### *3.2. SEM and EDS analyses of each biowaste*

Fig. 4 shows the results of SEM and EDS analyses before and after the column tests. The surface area of GS before the experiment was the smoothest compared with other filter media. Many protrusions were found on the surface of CS. On the other hand, more micro-pores were observed in WS compared with other biowaste filter media. The surface morphology of these media was evaluated as important property affecting adsorption of pollutant.

EDS analysis was performed to determine the chemical composition before and after experiment of biowaste demonstrated in Fig. 5. Carbon (C), oxygen (O), calcium (Ca) and magnesium (Mg) were analyzed as major constituents in the three biowaste filter media. After the experimental test run, the C composition increased by 5%–14% while

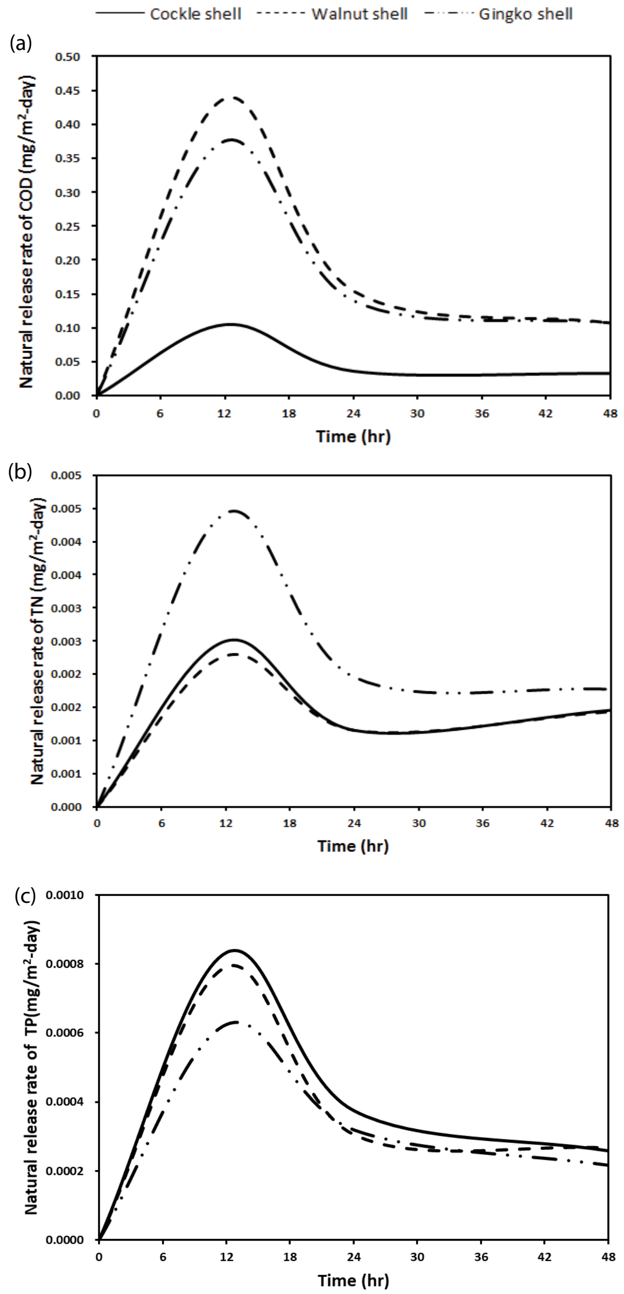

Fig. 3. Dissociation of unit mass of (a) COD, (b) TN, and (c) TP in natural filter media with respect to time.

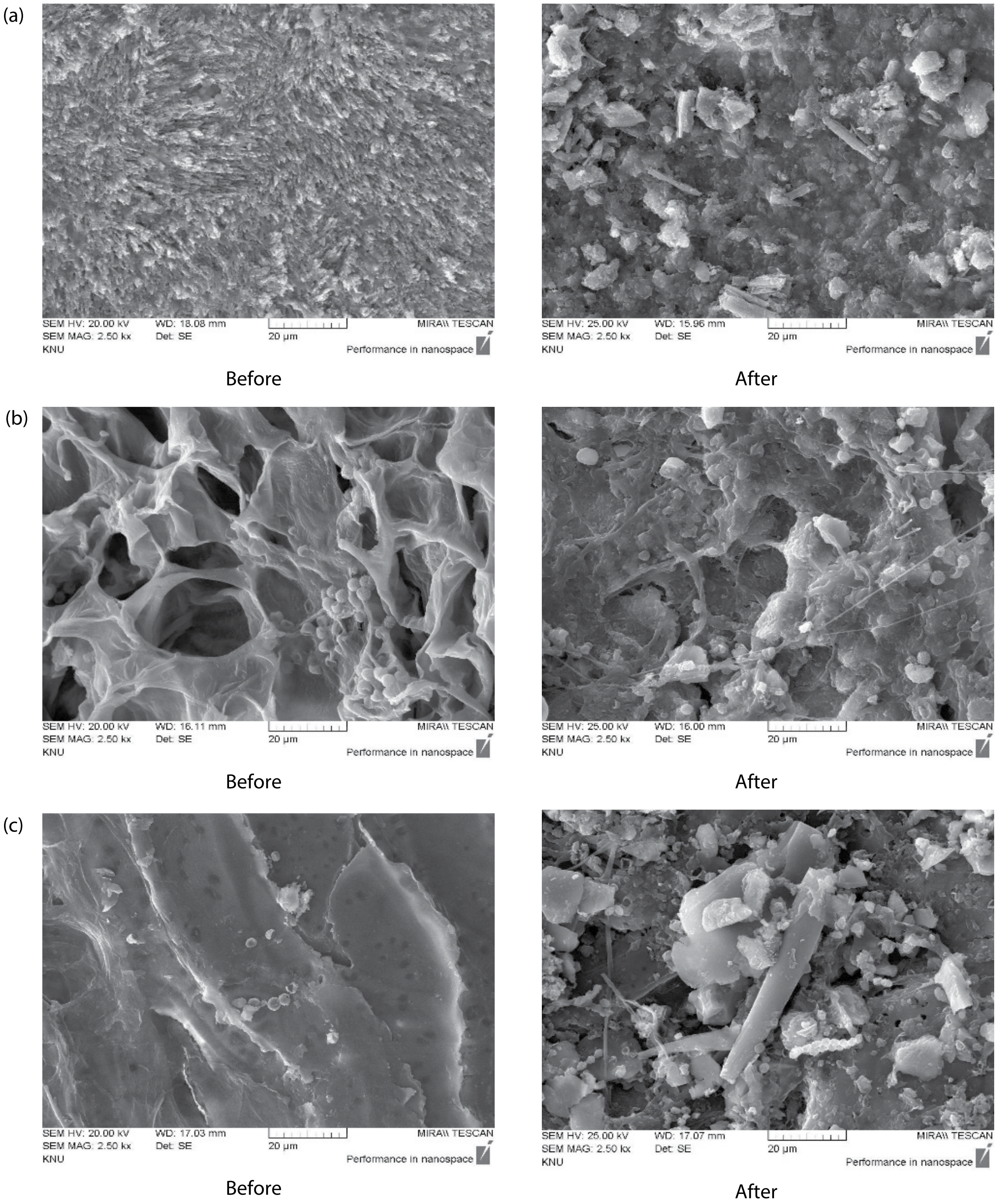

Fig. 4. SEM analysis of each natural filter media before and after column test runs for (a) cockle shell, (b) walnut shell, and (c) gingko shell.

O decreased by 12%–20% in all the biowaste filter media after the test runs. In addition, presence of aluminium (Al) and iron (Fe) were found after test runs. Unlike other filter media, sodium (Na) was found to be present in the CS because it is a marine waste. It is composed of $CaCO_3$ and calcium (Ca) composition ratio is relatively high. The composition ratio of C was increased about 8% after the experiment and the composition ratio was increased compared with other media such as Al and F. Silicon (Si) was observed in the shell of the shell before the experiment, and

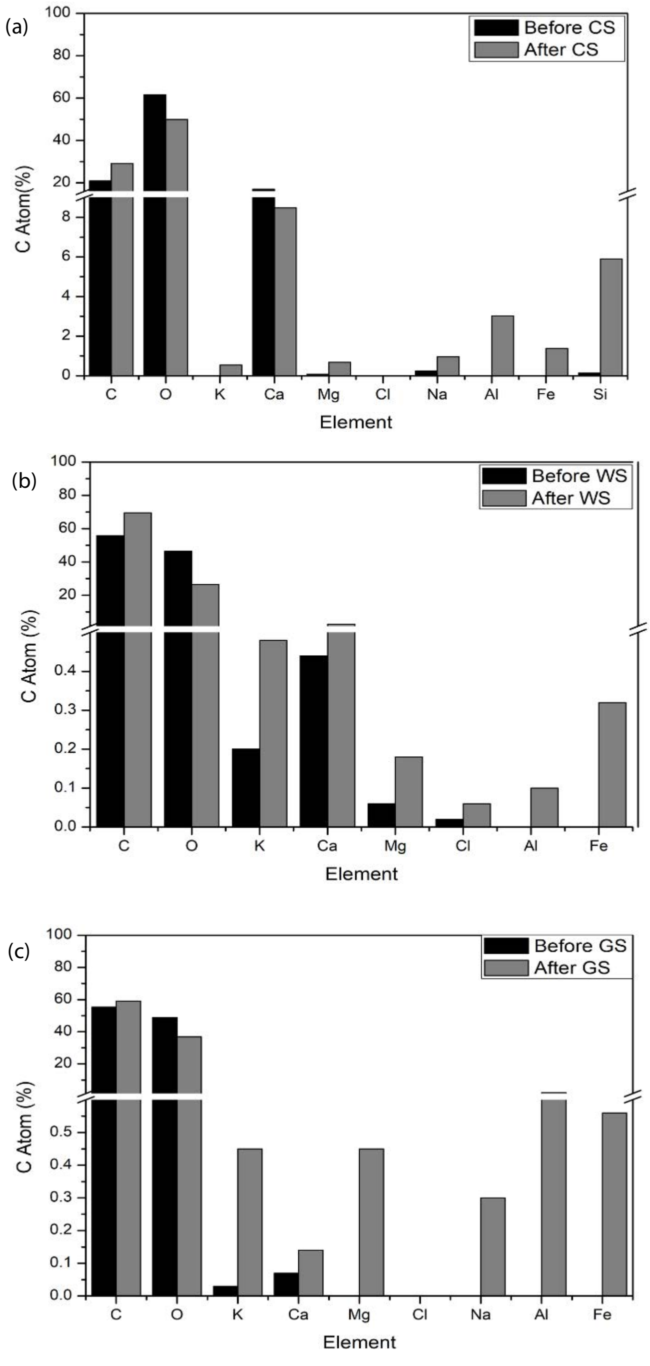

Fig. 5. EDS analysis of each biowaste filter media before and after column test runs for (a) CS, (b) WS, and (c) GS.

it was analyzed that the type and composition ratio of other heavy metals were increased compared with other natural filter media because of the high adsorption of heavy metals. CS, being a marine waste, was found to have Na unlike in other biowastes before the experiment. CS is composed of $CaCO_3$ which is the reason for high Ca content [20,21]. C, Al and Fe were increased by about 8%, 2.9% and 1.2% after the experimental test runs in CS. The micro-pores between the protrusions of CS and the micro-pores observed in WS contributed to the adsorption of trace contaminants. These findings revealed that both CS and WS were considered to be more effective in removing non-point pollutants containing high amount of minerals. Micro-pores will contribute to biological removal efficiency since it will serve as long-term microbial habitat [22].

### 3.3. *Evaluation of water balance and pollutant removal efficiency of biowastes by column test*

LID technologies are known to mimic the predeveloped state of an area by utilizing mechanisms including retention and infiltration through media porosity. Water balance of the biowastes filter media was exhibited in Fig. 6. Apparently, both cases exhibited greater than 87% infiltrated volume but it is evident that case 1 has higher retention by about 2%–6% due to smaller media particle sizes used. For both cases, it was also found that WS yielded 2%–8% greater retention compared with CS and GS which was due to the presence of both micro-pores and macro-pores on the surface of WS and the high moisture absorption capacity.

The pollutant removal of each biowaste is shown in Fig. 7. The reduction efficiency of TSS and COD of Case 1 for CS, WS and GS was greater than 70%. Smaller particle sizes of filter media implied more adsorption capability due to the increase in surface area. This led to higher reduction of pollutants by the biowaste [23,24]. TP showed more than 70% reduction CS and WS while GS showed less than 40% removal efficiency for Case 1. The presence of higher amount of $CaCO_3$ in the shell helped in increasing the P removal [25]. Low reduction of TN amounting to about 30% was observed in all filter media. This finding was attributed to short experimental period in which physical and chemical removal mechanisms were mostly optimized compared with biological removal mechanism.

Heavy metal removal was found to be greater in Case 1 compared with case 2 due to smaller particle sizes thereby producing larger surface area for filtration. Generally, urban stormwater runoff contains 120.1–422.1 µg/L, 94.9–403.7 µg/L and 11.1–32.0 µg/L Cu, Zn and Pb, respectively. The main heavy metals contained in the general urban stormwater runoff are Cu, Pb and Zn [26]. In the biowaste

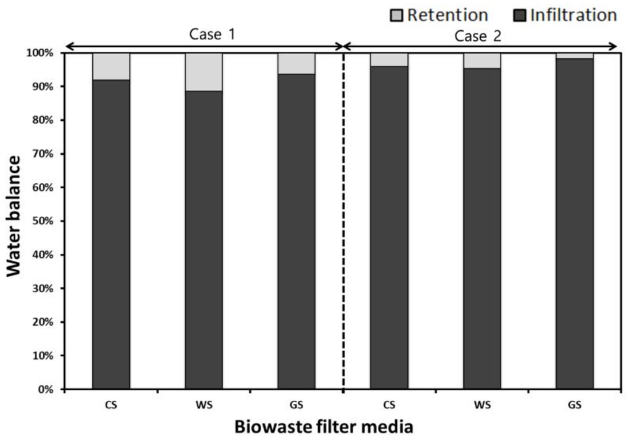

Fig. 6. Water balance.

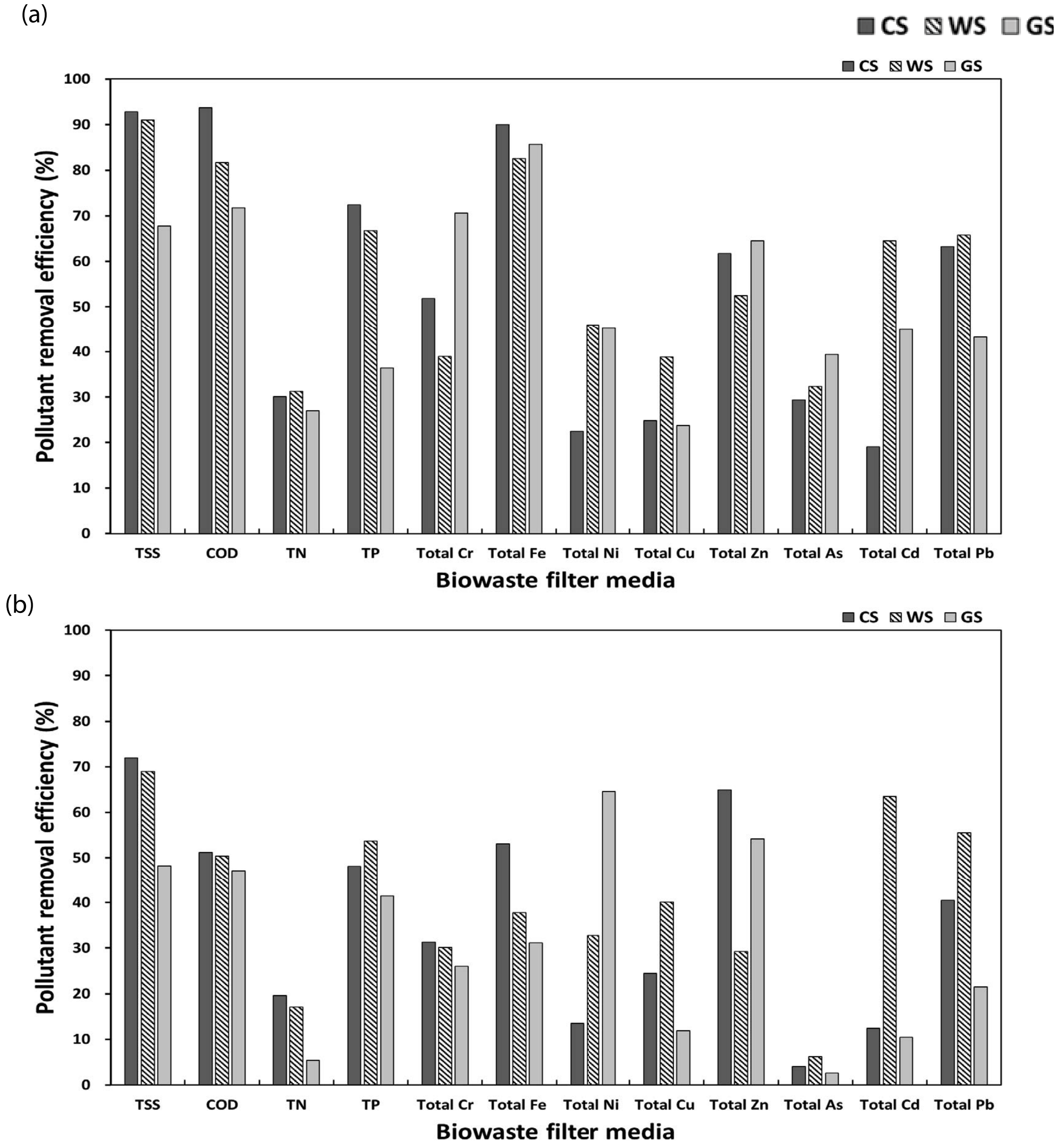

Fig. 7. Pollutant removal efficiency (a) Case 1 and (b) Case 2.

Table 4
Comparison of removal efficiencies of different filter media in different column studies

| Filter media type | Filter media particle size | Media depth | TSS | COD | TN | TP |
|---|---|---|---|---|---|---|
| | mm | cm | % | % | % | % |
| Sand(1) [27] | 0.17–0.3 | – | 96 | – | 11 | 85 |
| Sand(2) [27] | 0.3–0.84 | – | 96 | – | 1 | 10 |
| Zeolites [28] | 7–12 | 40 | 70 | 43 | 58 | 23 |
| Volcanic rocks [28] | 7–15 | 40 | 76 | 24 | 17 | 15 |
| Polyceramics [28] | 20–25 | 40 | 49 | 27 | 8 | 6 |
| Cockle shell | 50–100 | 35 | 82 | 72 | 25 | 60 |
| Walnut shell | 50–100 | 35 | 80 | 66 | 24 | 60 |
| Gingko shell | 50–100 | 35 | 78 | 59 | 16 | 39 |

filter media, the removal efficiency of Pb and Zn was about 40%–60%. To increase the removal efficiency of heavy metals, mixed application of the biowaste filter media with the inorganic filter media having high adsorption property is required. CS and WS showed high pollutant removal efficiency due to its micro-pores and larger surface area.

The comparison of the performance of different typical LID filter media to biowaste filter media is exhibited in Table 4. The TSS and TP removal efficiencies considering sand filter media were high but the removal efficiency of COD and TN was less than 11%. The removal efficiencies of TSS and TP in all three biowaste materials were 2%–16% higher than those of volcanic rock and zeolite. The removal efficiency of TSS, COD and TP in CS and WS which were typically through physical treatment mechanism was observed to be higher than GS due to the micro-pores and macro-pores formed on the surface of the surface of CS and WS. Therefore, it is analyzed that all biological waste resources are applicable as LID filter media.

## 4. Conclusions and recommendations

Application of LID techniques increased because of its high non-point source pollutant removal function and ability to restore the natural water cycle. Since urban storm water runoff contains various contaminants such as particulate matter, nutrients and heavy metals, it is necessary to apply various kinds of filter media in LID facilities. However, using filter media with high unit weight is not cost-effective specifically considering maintenance costs. This research evaluated the applicability of three biowastes such as CS, WS and GS which have lower unit weight than the typically used filter media such as sand or gravel in existing LID technologies. Based on the results of this study, the following conclusions may be drawn:

- Biowaste filter media have relatively smaller particle sizes compared with sand or gravel but have higher porosity and permeability coefficient implying that these are applicable as filter media of LID facilities. GS were found to require initial cleaning, as it showed the highest pollutant content. generated by the disruption of the membrane inside.
- Micro-pores in WS and CS increased the non-point source pollutant removal efficiencies of these biowastes. Micro-pores may be the long-term microbial habitat which will affect biological removal efficiency increasing the TN removal in the future.
- CS showed high infiltration and removal efficiency of pollutants, thus, it could be applied to vegetation type and infiltration LID facilities. WS may be applied to transportation land uses including parking lots and roads because of high heavy metal removal efficiency. In addition, it is applicable to constructed wetlands, bioretention, tree box filter and other vegetation type LID facilities due to high water retention and reduction of organic matter. GS was found to have the lowest pollutant removal efficiency but was assessed to be applicable to rooftops where low concentrations of non-point source pollutants were transported by stormwater and its properties including low infiltration rate and unit weight. It is considered that conjunctive use of organic and inorganic filter media will be more advantageous to be applied to LID technologies because the organic filter media was likely to settle due to oxidation during long-term use.

## Abbreviations

| | | |
|---|---|---|
| ADD | — | Antecedent dry day |
| CS | — | Cockle shell |
| $C(t)$ | — | Concentration |
| EDS | — | Energy dispersive spectrometer |
| EMC | — | Event-weighted concentrations |
| GS | — | Gingko shell |
| $Q_{TRu}(t)$ | — | Flow rate |
| SEM | — | Scanning electronic microscope |
| TSS | — | Total suspended solid |
| TN | — | Total nitrogen |
| TP | — | Total phosphorus |
| $Vol_{run}$ | — | Volume of runoff |
| $Vol_{dis}$ | — | Volume of discharge |
| $Vol_{infil}$ | — | Volume of infiltration |
| $Vol_{ret}$ | — | Volume of retention |
| $Vol_{sto}$ | — | Volume of storage |
| $Vol_{loss}$ | — | Volume of other lost water |
| WS | — | Walnut shell |

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