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The characteristics and performance of sustainable-releasing compound carbon source material applied on groundwater nitrate *in-situ* remediation

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HIGHLIGHTS

- SCCM developed from agricultural waste could sustainably release biodegradable carbon and applied to groundwater nitrate *in-situ* remediation.
- SCCM remediation system can achieve a chemical-biological denitrification process and remove 70–90% nitrate with little secondary pollution.
- This study demonstrated a novel utilization of straw-type agricultural waste as PRB filling matrix for groundwater nitrate in-situ remediation.

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ABSTRACT

Recently, reductant and carbon source were frequently used in groundwater nitrate remediation worldwide. Agricultural waste as a promising organic carbon source, has been paid much attention but the problem of sustainability, bioavailability and secondary pollution remained unsolved. This study was conducted to depict the characteristic and performance of developed sustainable-releasing compound carbon source material (SCCM) applied on the *in-situ* remediation of nitrate in shallow groundwater. Results showed the SCCM based on agricultural waste and zero valent-iron (ZVI) has a stable carbon releasing rate, which is suitable for stimulating the low microbial active environment in groundwater continuously, and capable of avoiding rapid TOC releasing in the early stage. The released carbon sources in SCCM leachate were mainly small molecular alcohols and acids with high microbial availability. As *in-situ* permeable reactive barrier (PRB) filling material, SCCM can form an optimal carbon source radiation range of 20 cm, with a maximum efficient carbon source radius of 1 m, which can reach an extended active zone. A positive correlation between the ZVI content and nitrate removal rate was found. The chemical and microbiological evidence both indicated that the expected chemical reduction and biological denitrification was gradually established. Additionally, the absorption of ammonia and chroma by attapulgite effectively avoided the secondary pollution. In conclusion, the application of SCCM in groundwater nitrate *in-situ* remediation optimized the nitrate removal efficiency and provided theoretical basis for engineer carbon sources development from straw-type agricultural waste.
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1. Introduction

The nitrate pollution in groundwater is widespread, which is mainly caused by the excessive nitrogenous fertilizer application in agricultural production. In recent years, many researches and engineer practice had been conducted on *in-situ* remediation of nitrate pollution in groundwater. The technologies most widely used are chemical-biological denitrification method, which involves adding reductant and carbon source to stimulate chemical and biological reactions.

ZVI has also frequently used as chemical reductant in the nitrate removal from groundwater. Beside the chemical reduction, the H_2 released from ZVI corrosion also act as an electron donor and promote the propagation of hydrogen autotrophic denitrifiers (Son et al., 2006; Yu et al., 2007). However, ZVI is susceptible to solution pH, ion composition and other factors, which can result in precipitation, soil agglomeration, and non-conducive to of nitrate

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reduction. Besides, ammonium remained in the system as the main product of nitrate reduction by ZVI, which did not achieve the goal of complete removal of nitrogen in groundwater (Lu et al., 2017).

It has been found that bio-process can be potentially activated by carbon source, and both nitrate and toxic nitrite in the groundwater can be effectively removed (Lan et al., 2015). Therefore, the nature of carbon sources played a key role in the nitrate removal efficiency (Li et al., 2012). If plenty of organic carbon was available, it can use by some microbial strains as electron donor in the denitrification reaction (Qin et al., 2017). However, if groundwater lacks organic carbon, heterotrophic denitrification could be limited (Sunger and Bose, 2009). Denitrification, considered as the primary nitrate removal mechanism in groundwater, comprises heterotrophic denitrification (Capodaglio et al., 2016) and autotrophic denitrification (Molognoni et al., 2017). Heterotrophic bacteria use organic matter as electron donor, and nitrate act as electron acceptor instead of dissolved oxygen. In autotrophic denitrification, bacteria utilize carbon and inorganic (H_2, iron species) as electron donor. Therefore, many nitrate removal processes have been developed for each kind of electron donor (Park and Yoo, 2009), among which organic carbon was the most economic and efficient one.

At present, the carbon source used for nitrate removal were mainly divided into organic carbon compounds and agricultural waste. Organic carbon compounds include methanol, ethanol, acetic acid, glucose, etc., which were investigated to achieve higher denitrification levels and rates via biological process (Sahinkaya et al., 2011; Sahinkaya and Dursun, 2012; Qambrani et al., 2013). But this kind of carbon source was supplemented and costing, which may also pose potential environmental pollution under incomplete reaction.

Agricultural waste has been demonstrated as an alternative to organic carbon compounds for nitrate treatment, since the efficient utilization of which reduce the cost and negative environmental impacts. Agricultural waste such as cotton, wheat straw, rice husk, bark, corn, sawdust, wood chip, rice straw have been found effective to provide carbon source for denitrifying microorganisms. Sawdust or wood chip as the most commonly used substrates, are successfully applied in field-scale projects (Schmidt and Clark, 2012; Li et al., 2016a,b). The biological denitrification processes have persistence (5–15 years), high nitrate removal rates ($1–20\ g\ N \cdot d^{-1} \cdot m^{-3}$media), and low maintenance costs (Robertson, 2010a). However, agricultural waste does have a drawback though. Excessive ammonium is released as an intermediate (Yang et al., 2012) of dissimilatory nitrate reduction (Zhang et al., 2012). Furthermore, most of the agricultural waste have uncontrolled carbon release and slower starting of denitrification. Some adverse effects such as high content of colored organic matter in the effluent and accumulations of nitrite (NO_2^-) can be observed. Therefore, more attention should be paid on the effective utilization and elimination of secondary pollution of agricultural waste carbon sources (Chang et al., 2016). Chemical reduction methods can quickly start the reaction system at the beginning, and biological denitrification methods is advantageous to long and sustainable remediation in the middle and later stage. If combined, they will have an expected application prospect. Wang found that the combined addition of sawdust and ZVI had better denitrification effect than individual (Wang et al., 2008). Oh investigated that nitrate can be effectively denitrified by microbes in natural and engineered systems by mixed cultures in the presence of ZVI (Oh et al., 2016).

In this research, agricultural waste and ZVI were used as the raw material with a mixtures of mineral materials and plastic materials to develop series of high-performance SCCM, whose characteristics including carbon releasing, biodegradability effectiveness, secondary pollution, chroma and soil agglomeration elimination ability, as well as nitrate removal performance were verified with batch and pilot experiments. Furthermore, the engineering parameters of developed SCCM used as PRB filling matrix were optimized. This practice may achieve the feasibility of straw-type agricultural waste as carbon source and certify the effectivity of chemical-biological denitrification method in groundwater nitrate *in-situ* remediation.

2. Materials and methods

2.1. Material composition and preparation

SCCM was developed with uniform mixing rolling granulation process taking agricultural waste and ZVI as raw material, mineral materials, plastic materials and viscous materials as accessories. SCCM were produced according to preferred parameters including particle strength, bulk density, density, carbon content, price and other factors. The specific requirements were as follows: 1) high particle strength, 2) density >$1.0\ g\ cm^{-3}$, not easily expandable and disintegrate in the water, 3) high denitrification efficiency, and 4) ideal carbon releasing sustainability.

SCCM was composed of core and shell as shown in Fig. 1. The core act as carbon source provider, which was consisted of agricultural waste, ZVI and other raw materials. The shell was composed of attapulgite and other mineral materials, which provide porous structure to effectively improve the organic carbon releasing and the reducibility of ZVI, and carrier for microbial attachment. In particular, the attapulgite clay, both used in cores and shell, was a binder to consolidate SCCM, as well as absorbent for ammonia.

2.2. Experiment design

2.2.1. Carbon source releasing test

The experiment was conducted to investigate their carbon (TOC) releasing performance and kinetics in the simulated groundwater. Six SCCM based on wheat straw, corn stalk, corn cob, wood chip, rice husk, sugar cane, and the corresponding agricultural waste were tested.

50 g of each SCCM and raw material were preserved in 1 L blue bottles, respectively. About 500–900 mL of distilled water was

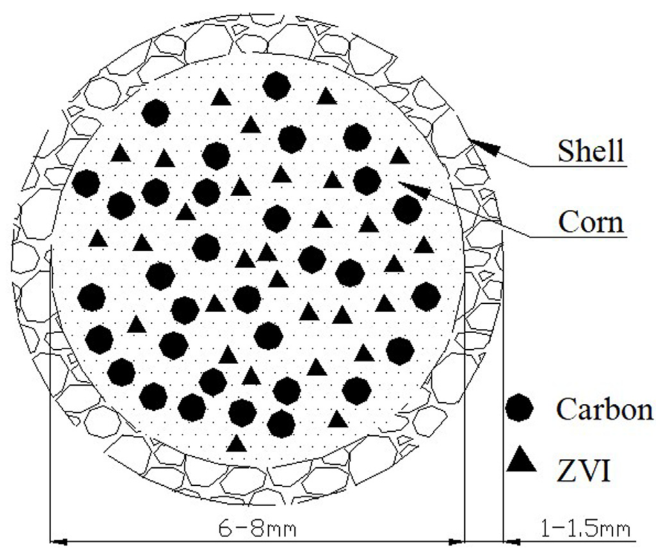

Fig. 1. Schematic structure of SCCM.

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