



## Full Length Article

# Gaseous emission and ash characteristics from combustion of high ash content antibiotic mycelial residue in fluidized bed and the impact of additional water vapor

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## ABSTRACT

The gas (mainly NO and SO<sub>2</sub>) emission and ash characteristics from combustion of high ash content antibiotic mycelial residue (AMR) rich in N and S were investigated using a laboratory scale fluidized bed combustor, and the effects of addition of water vapor studied at the same time. The tested combustion temperature, excess air ratio ( $\alpha$ ) and mass ratios of water vapor-to-fuel (WV/F) varied in 750–950 °C, 1.3–1.9 and 0.4–1.4, respectively. Without additional water vapor present, both the concentrations of NO and SO<sub>2</sub> in flue gas increased with elevating combustion temperature and excess air ratio, corresponding to the increases in conversion ratio of fuel-N to NO and emission ratio of S. The addition of water vapor facilitated excavating organic matters in the ash to result in its lower C and N contents. Additional water vapor formed partial reducing atmospheres, which were characterized by high CO and H<sub>2</sub> concentrations in combustion atmosphere and low valence S in the ash, to thus visibly reduce NO. The SO<sub>2</sub> concentration became slightly lower at the presence of additional water vapor and further decreased with more water vapor addition. Additional water vapor strengthened capture of SO<sub>2</sub> by the ash. Higher WV/F ratios led to more porous structures in ash, accountable for NO reduction by catalytic effect as well as SO<sub>2</sub> absorption. Nevertheless, it seems less possible to reduce NO and SO<sub>2</sub> concentrations to meet their respective emission criteria, and other measures will thus have to be taken to control their emissions when directly combusting AMR for energy recovery.

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## 1. Introduction

Annually, millions of tons of antibiotic mycelial residue (AMR) are generated in China – the leading producer of antibiotics in the world [1]. As one of the fermentative process wastes, various AMRs mostly consist of mycelia and fermentative substrates and are rich in moisture (usually over 80 wt%). Moreover, the water in these kinds of biowastes mainly exists as various “bound” forms and is difficult to be mechanically removed [2]. The AMRs are thus easy to cause serious environmental pollution if discharged without prior innocent treatment, especially considering that certain amounts of antibiotics unavoidably reside in them. Above all, the residual antibiotics have been verified to bring about great risks of food chain safe for animal and human being (e. g. drug resis-

tance) [3]. As a result, these AMRs have been listed on the category of “hazardous wastes” in the recently updated Chinese environmental laws, which prohibit open disposal and use of them as animal feedstuff, land fertilizer or similar usage. Unfortunately, most conventional technologies such as biochemical methods are out of work for treatment/disposal of the AMRs because the residual antibiotics greatly inhibit the microbes frequently present in the biochemical treatment systems, and up to this day no effective methods are available. Therefore, feasible and effective technologies are in urgent need to treat/dispose these AMRs.

On the other hand, the AMRs rich in organic matters surely fell within the scope of biomass resource. Generally, combustion is very effective for safe disposal of bio-wastes and simultaneous recovery of their bio-energy. Among various combustion techniques widely used for burning biomass for heat and power, fluidized bed combustion with its high efficiency is most suitable for wet material due to partial gasification of fuel prior to combustion [4–6]. Therefore, fluidized bed combustion technology was

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employed here to combust AMRs, to realize their ultimate disposal and energy recovery.

Similar to most industrial biomass resources, the AMRs are rich in nitrogen and sulfur. Attention should, thus, be paid to high  $\text{NO}_x$  and  $\text{SO}_2$  concentrations in flue gas from AMRs combustion. The  $\text{NO}_x$  emission from combustion of nitrogen-rich biofuel and its corresponding control methods have been extensively studied and developed. Especially, more and more emphasis was recently attached to the NO emission characteristics from biofuel combustion with addition of water vapor [7], aiming to perform direct combustion of biofuels usually having high moisture contents and thus avoid biofuel drying with high energy consumption. The positive effects of additional water vapor on NO reduction have showed attention by other researchers [8–13]. The roles of additional water vapor present in the combustion atmosphere were argued, such as diluting oxygen concentrations and lowering flame temperatures to inhibit NO formation [8], strengthening NO reduction by CO [9] via hindering CO oxidation [10]. Li et al. [11] found that higher water vapor contents and temperatures (below 1100 °C) facilitated NO reduction and argued that the NO reduction rested with the combined effect of hydrogen free radical (H $\cdot$ ) and CO. Generally, less NO emission was closely related to generation of more CO and H $_2$  (or H $\cdot$ ) with sufficient water vapor present in the combustion atmosphere [12,13]. By employing fluidized bed to combust sewage sludge (similar in composition and structure to AMRs) with different moisture contents, several researchers observed that higher moistures did result in lower  $\text{NO}_x$  emission concentrations [14,15]. Based on the above results, it is expectable to reduce NO emission to certain degrees when performing fluidized bed combustion of high water content AMRs for cleaner energy recovery. In comparison with a number of studies on NO emission from combustion of high nitrogen content biofuels at the presence of water vapor, few literatures cover the  $\text{SO}_2$  emission from combustion of high sulfur content biofuels, especially in water vapor rich atmospheres.

Indeed, the AMRs have high ash contents. Little knowledge can be obtained about comprehensive gaseous emission characteristics from combustion of this kind of biofuel. For the purpose of clean energy recovery of the AMRs via combustion, it is necessary to systematically investigate the  $\text{NO}_x$  and  $\text{SO}_x$  emissions and the impact of water vapor. Above all, the interactions between gas products, ash and water vapor need to be deeply revealed. This leads to this work.

## 2. Experimental section

### 2.1. Fuel

The tested AMR was the process residue from cephalosporin C (CPC) production by fermentation with corn starch, bean protein and other additives as the substrates, and was supplied from a pharmaceutical factory in Hebei province, China. The moisture content of the raw AMR is as high as about 83 wt%. For performing combustion tests, the AMR was subjected to air drying in an electric oven at 110 °C for 24 h and was in turn ground and screened to the size of 0.2–0.5 mm. The results of proximate (according to the Chinese standard (GB/T 28731-2012)) and ultimate analyses of the prepared dry sample are shown in Table 1. The dry AMR contains 1.90 wt% nitrogen, which is comparable to those in extremely high nitrogen coals. The total S content of the dry AMR is high as 3.46 wt %, remarkably higher than normal biomass and most coals. The high nitrogen and sulfur contents mean a latent risk of emission of gas pollutants such as  $\text{NO}_x$  and  $\text{SO}_2$  when the AMR is combusted for safe disposal or energy recovery. The two ash samples were prepared to characterize the ash properties. The first one (No. 1

**Table 1**

Proximate and ultimate analyses of the tested antibiotic mycelial residue (dry base, wt%).

Analysis	Value
Proximate Analysis	
Volatile	64.01
Ash	34.99
Fixed Carbon	1.00
Ultimate Analysis	
C	42.76
H	5.37
N	1.90
Total S	3.46
O <sup>a</sup>	11.52
HHV <sup>b</sup> (MJ/kg)	19.27

<sup>a</sup> By difference.

<sup>b</sup> High heating value.

ash) was obtained by combusting the AMR stacked in a closed muffle furnace gradually heated to 900 °C (at around 20 °C/min) to capture the generated  $\text{SO}_2$  to the maximum degree (with maximum  $\text{SO}_2$  absorption). The second one (No. 2 ash) by combusting the AMR scattered in a preheated (900 °C) tube furnace at air flow atmosphere to sufficiently emit the  $\text{SO}_2$  (with minimum  $\text{SO}_2$  absorption). Table 2 gives the compositions of the two ash samples. For the No. 2 ash with minimum  $\text{SO}_2$  absorption, its most main composition is CaO with such a high content as 50.22 wt%, and the second one is  $\text{P}_2\text{O}_5$  (24.81 wt%) followed by  $\text{SO}_3$  (18.70 wt%). In comparison, for the No.1 ash with maximum  $\text{SO}_2$  absorption, although CaO is still the most main composition (42.20 wt%) the  $\text{SO}_3$  content is as high as 32.36 wt%, higher than that (21.14 wt%) for  $\text{P}_2\text{O}_5$ . The deformation temperature of the No. 1 ash is above 1500 °C, so agglomeration is not expected to occur when the AMR is combusted at lower temperatures.

Though the total S content in the AMR is extremely high (3.46 wt%), the organic S content in the AMR is just 0.08 wt%, according to the results of speciation analyses of sulfur in No. 2 ash which are showed in Table 3. Almost all the sulfur in the ash appeared as sulfates. The total S and sulfate sulfur contents in the ash were 7.10 and 7.08 wt%, respectively. Considering the high ash content (ca. 35 wt%) of the AMR, part of sulfate sulfur was surely converted into sulfurous gases (mainly  $\text{SO}_2$ ) in addition to organic sulfur, which was explainable for high  $\text{SO}_2$  concentrations from the subsequent combustion tests (refer to Section 3.3).

### 2.2. Apparatus and method

All the combustion tests were carried out in a stainless steel fluidized bed combustor shown schematically in Fig. 1. The combustor mainly consists of a fluidized bed with two combustion chambers respectively equipped with two outer electrical heating furnaces, and the lower and upper ones were expected to perform combustion of the solid fuel and the produced volatile, respectively. The two combustion chambers have the same inner diameter of 68 mm, but the height is 740 for the lower one and 640 mm for the upper one. The lower combustion chamber is connected to a screw feeder, a water cooler, a gas supply system and a vaporizer. The gas supply system is composed of Air,  $\text{N}_2$ ,  $\text{CO}_2$ ,  $\text{O}_2$  cylinders assembled with a gas mixer and a gas distributor. The water vapor supply system is composed of a constant-flux pump and a vaporizer. The upper combustion chamber is connected to a cyclone, a condenser system (the water cooled condenser and an ice condenser), a flue gas analyzer and a data acquisition system.

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