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# Hydrothermal gasification of sewage sludge and model compounds for renewable hydrogen production: A review



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## ABSTRACT

Sewage sludge is bio-solid with high moisture content generated from wastewater treatment plants. Due to the avoidance of energy-intensive dewatering, hydrothermal conversion of sewage sludge becomes a promising technology to simultaneously achieve energy recovery and solid waste management. In order to obtain an entire understanding of applicability of hydrothermal gasification for hydrogen rich gas production from sewage sludge, this review article discussed hydrothermal conversion and gasification processes in terms of fundamental principles, operating conditions, partial oxidative gasification, and detrimental effects of intermediates. Furthermore, since organic compounds in sewage sludge are mainly composed of carbohydrates, proteins, lipids, and lignin, this article comprehensively reviewed hydrogen production from these biomass model compounds and their hydrolysis products under sub- and supercritical water. Additionally, introduction of alkali salts and heterogeneous catalysts to enhance hydrogen yield under mild temperatures and pressures in hydrothermal gasification process was also discussed. Based on bench and pilot scale studies, supercritical water gasification of sewage sludge for hydrogen production is feasible in terms of technical and economic evaluation. Given issues concerning corrosion, plugging and high operating cost, a combined supercritical water gasification and catalytic hydrothermal gasification concept is proposed as a practical strategy to directly harness hydrogen from sewage sludge in future applications.

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## 1. Introduction

Sewage sludge is a semi-solid residue generated from wastewater treatment plants (WWTPs). Wastewater treatment system is generally integrated with a series of physical, chemical, and biological treatment units. After anaerobic digestion, the sludge is subjected to conventional mechanical dewatering which can reduce the moisture content to around 80 wt%. Sewage sludge is rich in organic matter and nutrients (such as nitrogen and phosphorus) and mainly composed of proteins (~40%), lipids (10–25%), carbohydrates (14%), lignin and ash (30–50%) [1,2]. Depending on the characteristics of wastewater sources and wastewater treatment process, heavy metals (i.e. Cd, Cr, Cu, Zn, Pb, Ni, and As), pathogens or other microbiological pollutants, persistent organic pollutants (POPs) (i.e. polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), dioxins, pesticides, nonyl phenol, and linear alkyl sulfonates, etc.) [3] can be entrained in sewage sludge, posing potential risks to environment and human health if it is not appropriately treated.

Due to urbanization and increasingly stringent regulations for the quality of discharged water, the sludge generation from WWTPs is soaring worldwide. At present, the annual production of dry sludge in European Union is more than 10.96 million tons [4] and sludge production in China is estimated to reach 6 million dry tons per year [5]. Moreover, the cost used for sludge treatment represents almost 50% of the total capital cost of wastewater treatment process [6]. Therefore, a wide variety of sludge treatment technologies have been implemented, including three main categories, landfill disposal, land application, and incineration.

Landfill disposal not only occupies plenty of land but also generates a large volume of landfill leachates and gases resulting from the high moisture and organic matter contents in sewage sludge. The landfill leachates and gases can incur pollutions to surrounding water, air, and soil. Sometimes, the landfill site may even collapse due to insufficient strength of sludge. Moreover, metabolites in the leachates can be more toxic than its precursors [7]. Composting is a prominent technology for land application. However, the odor control is problematic both in the processing stage and storage areas [6]. Oleszczuk [8] reported composting could significantly increase the toxicity of sewage sludge, which may be associated with the increased mobility and bioavailability of metabolites [7]. On the other hand, the market for composted sludge is a prerequisite. Thus, suitable revenue for the composting facilities is required and more outlets for the final compost should be developed [6]. Given migrations of heavy metals and organic

pollutants, it may not be advisable to use sewage sludge derived compost for land application. Furthermore, due to the high calorific value in sludge (i.e. higher heating values for dry sewage sludge before and after digestion around 23 MJ/kg and 8.5–17 MJ/kg, respectively), energy recovery from sludge has attracted great attention recently [9]. The potential energy in sludge could be recovered using either biological or thermochemical processes. In particular, incineration can dramatically reduce the sludge volume. Nonetheless, before being fed into incinerator, the moisture content of sludge should be lowered down to 35% during pre-drying step, leading to high energy consumption [7]. To eliminate the energy-intensive drying process, hydrothermal conversion technology was introduced to recover energy from wet biomass waste.

Hydrothermal conversion is a thermochemical process to reform biomass in hot compressed water. Under elevated temperature and pressure, specifically when exceeding the critical point (374.3 °C and 22.1 MPa) of water, the density, static dielectric constant and ion dissociation constant of water drop drastically, which can accelerate the reaction rate substantially [10]. Due to those superior properties of hot pressurized water, it acts as a non-polar solvent and benign reactant with high diffusivity, excellent transport properties and solubility. Consequently, hydrothermal conversion technology has been widely applied for fuels and chemicals recovery from wet biomass and organic waste with high moisture content in the last two decades. As shown in Fig. 1a, hydrothermal conversion can be divided into (1) hydrothermal carbonization (180–250 °C) for hydrochar production [11,12], (2) hydrothermal liquefaction (about 200–370 °C, with pressures between 4 and 20 MPa) for heavy oil production and (3) hydrothermal gasification (near-critical temperatures up to about 500 °C) [13] to generate hydrogen rich gas under various conditions. From the perspectives of fossil energy shortage and environmental impacts, renewable hydrogen recovery from readily available wet biomass using hydrothermal gasification is desired in the long run. Kruse [14] summarized previous research work relating to supercritical water gasification (SCWG) of biomass without heterogeneous catalysts whereas Elliott [15], Guo et al. [16] and Azadi et al. [17] reviewed catalytic sub- and supercritical water gasification of biomass for hydrogen production. In recent years, more efforts have been made to explore strategies to enhance hydrogen yield and carbon gasification efficiency during hydrothermal conversion of biomass or wastes. Up-to-date information and comprehensive summary are imperative to highlight technical and economic feasibility of hydrothermal gasification technology for wet biomass waste management system, especially

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