A Review on Sewage Sludge Valorization via Hydrothermal Carbonization and Applications for Circular Economy

Dilvin Çebi
Ege University: Ege Universitesi
Melih Soner Celiktas (soner.celiktas@ege.edu.tr)
Ege University Solar Energy Institute https://orcid.org/0000-0003-0597-5133
Hasan Sarptaş
Ege University: Ege Universitesi

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Abstract

In pursuit of establishing a circular economy, waste-to-energy approach is gaining increasing attention. In this manner valorization of sewage sludge constitutes a critical importance due to generation in high quantities, difficulties in disposal and associating environmental impacts. Hydrothermal carbonization (HTC) is a relatively recent, however acclaimed method for sewage sludge management and valorization due to process compliance with sludge characteristics.

In this review, research studies are evaluated under the categorization of application fields of sludge derived HTC products such as solid fuel production, gas production, soil remediation, nutrient recovery, water treatment and energy storage. Research findings are compiled and a network mapping is employed for the visualization of the current situation and correlation in respective fields. The potential of HTC for sewage sludge valorization and future projections concerning available techniques are assessed within the context of circular economy.

Introduction

Industrialization and population growth led to increasing quantities of sewage sludge raising environmental concerns (Wang et al. 2018)(Zhai et al. 2014)(Zan et al. 2020)due to contaminants, pathogens and toxic elements in its composition (Gherghel et al. 2019). Sludge management having a high energy requirement refers to 20–60% of total operation costs of wastewater treatment plants (WWTPs)(Gopinath et al. 2021). The environmental impacts are much severe considering 50% of greenhouse gas (GHG) emissions from WWTPs are attributable to sludge disposal (Brown et al. 2010) making sludge valorization a pressing issue.

Realizing a sustainable development is possible by adopting a circular economy approach (Emmanuel et al. 2021) keeping resources, product and materials at their highest value (Caneghem et al. 2019)(Barros et al. 2020) rendering the conversion of waste to energy inevitable(Sharma et al. 2020). In this manner, the regenerative system implied within the circular economy concept (Sikdar 2019) denoting waste minimization and reuse through conversion of waste to energy (Anaurbekovna et al. 2019) (Sharma et al. 2020)is adaptable for valorization of sludge by currently available technologies.

Evaluation of sewage sludge as a valuable source rather than a waste material to be disposed of will increase energy and material recovery via clean technologies such as hydrothermal carbonization (HTC). Differing from other thermochemical processes, hydrothermal carbonization emerges as a promising method for sludge management primarily due to its ability to process wet materials without pre-drying (Escala et al. 2013)(Mäkelä et al. 2016)(Ahn et al. 2020)(Pecchi et al. 2020). Operating at relatively low temperatures of 180–250° (Merzari et al. 2019) and short retention times (Gai et al. 2016a)(Chen et al. 2020a) HTC is a cost effective process (Chen et al. 2020b) for converting sewage sludge to carbonaceous material (He et al. 2013) (Wang et al. 2018) with several application options such as solid fuels, adsorbents and fertilizers (Singh et al. 2020).

These options include utilization of hydrochar produced via HTC as a solid fuel. HTC process is proven to enhance hydrochar properties in means of heating value close to that of lignite (Danso-Boateng et al. 2015), fuel ratio and aromatic structure (Reza et al. 2014). Another favored application field of hydrochar is for soil remediation and plant growth (Xu et al. 2018), promoting carbon sequestration (Malghani et al. 2013) (Naisse et al. 2015).

Hydrochars are also used in wastewater treatment as adsorbents due to their characteristics in relation to surface area, porous texture and mineral composition (Wang et al. 2018). Sewage sludge based hydrochars show higher efficiency in removing dye and heavy metals from waters compared to commercial activated carbons (Singh et al. 2020).

Additionally, the fact that the liquid phase of HTC process (LFHTC) is also being assessed in recent research studies (Nyktari et al. 2017)(Villamil et al. 2018)(Villamil et al. 2019)(Xu et al. 2020) as feeding material due to high organic and nutrient content is encouraging in pursuit of circular economy.

Hydrothermal Carbonization Of Sewage Sludge

The applications of products obtained from the hydrothermal carbonization of sewage sludge are evaluated in detail under following categories: (i) solid fuel production; (ii) gas production; (iii) soil remediation; (iv) nutrient recovery; (v) water treatment and (vi) energy storage.

Solid Fuel Production

Xu et al. (2019) treated sewage sludge and cow dung biogas residue by co-HTC. Higher carbon content and higher heating value (HHV) of the hydrochars are observed when the two materials are carbonized together. Addition of biogas residue had a significant effect on the hydrochar combustion quality.

Wilk (2016) conducted HTC experiments of municipal sewage sludge at 200° and different retention times (4, 7, 10, 12 h) followed by the characterization of hydrochars produced to be used as a solid fuel. The optimized retention time is determined as 4 h in line with the ash content of the hydrochars (Wilk 2016).

To propose a new method for sewage sludge treatment Villamil et al. used HTC for hydrochar production from sewage sludge, coupled with AD of primary sewage sludge and the process water of HTC. The minimum COD removal rate is reported as 53%. The energy recovery rate is 41.5%. Emphasis is made to further modelling and design for large scale HTC plants (Villamil et al. 2020b).

Liu et al. performed hydrothermal carbonization and oxy-fuel combustion of sewage sludge for examining the combustion properties. Combustion kinetics of sewage sludge derived hydrochars by HTC indicates the high quality of these solid fuels. HTC process also improved the dewaterability of sewage sludge, resulting in a 30% energy save for drying (Liu et al. 2019). It is further reported that HTC prevents problems related to ash formation.

Combustion properties of hydrochar produced from the hydrothermal carbonization of sewage sludge at 200° is investigated in the study by He et al. (2013). Requiring lower activation energy, with high carbon and low volatile matter content, hydrochars are found to possess a much higher combustion quality than the sewage sludge. Due to a removal rate of 60% of nitrogen and sulfur hydrochars produced can be considered a more environmentally safe option (He et al. 2013). The energy recovery aspect is also found noteworthy due to the avoided step of drying.

Fakkaew et al. (2015) conducted a “two-stage HTC” experiment consisting of hydrolysis and carbonization processes to compare the effects on hydrochar production from faecal sludge. As a result the two-stage process was favored in means of energy efficiency over conventional HTC (Fakkaew et al. 2015).

In another study using the same feedstock Fakkaew et al. investigated the characteristics of hydrochar produced from the HTC of faecal sludge. Based on the hydrochar properties it is suggested that the products with energy values of 19–20 MJ/kg matching to coal (Fakkaew et al. 2018), could be suitable for several applications while drawing attention on the necessity for an economic feasibility study for the development of HTC technology on a larger scale.

Zhai et al. used different biomass to hydrothermally carbonize sewage sludge for hydrochar production, taking into account the energy consumption for the evaluation of method and material feasibility. Different biomass including sawdust, corncob, corn stalk and rape straw was used with sewage sludge for HTC. The most efficient biomass addition was selected as corncob. The optimized operation condition is determined as 300°, 60 min and the energy recovery rate is reported as 71.60% (Zhai et al. 2017).

A comparison on the effects of pine and acacia additions to the hydrothermal carbonization of sewage sludge Wilk et al. experimented at 200°, 4 h. The results proved that the energy values of lignocellulosic biomass increased with HTC making it a preferable method in means of hydrochar production to be used as solid fuel (Wilk et al. 2019).

Kim et al. optimized the temperature of HTC at 220° for the sewage sludge sample for solid fuel production. The calorific value of the hydrochar produced was comparable to that of coal (Kim et al. 2014), suggesting the desirability of HTC for solid fuel production from sewage sludge.

Parshetti et al. experimented on the co-combustion of hydrochar produced from sewage sludge at 250°, 15 min retention time with raw coal and upgraded coal. The hydrochar is reported to have 15.82 MJ/kg energy content and the co-combustion with raw and upgraded coals while enhancing the combustion quality also reduced the emissions of CO_2, CO and CH_4 (Parshetti et al. 2013).

Lee et al. (2019) produced hydrochars through the co-HTC of primary sludge and *Chlorella sp.*, demonstrating that the hydrochar properties benefited from the cultivation of the biomass resource addition. Energy content of 24 MJ/kg and low ash value is found suitable for hdyrochars to be used as solid fuel (Lee et al. 2019).

Co-HTC experiments of sewage sludge and lignite were conducted by Song et al. at 120° – 300° and observed an upgrade in calorific value, fixed carbon and carbon content of hydrochars at the optimal condition of 240°, at 1:1 ratio (Song et al. 2019). The interactions of the two feedstocks demonstrated a more stable combustion with a lower activation energy requirement, indicating the use of hydrochars as a clean fuel alternative.

He et al. diversified the co-HTC of sewage sludge with the addition of fruit and agricultural wastes such as rice straw, peanut shell, orange peel and leaves to examine the fuel properties of hydrochar. For all feeding material at different loading and mixing rates the energy content of hydrochars were increased. The highest energy content of hydrochars was reported as 21.72 MJ/kg for peanut shell and sludge mix at 3:1 ratio (He et al. 2019) while combustion characteristics of hydrochars presented a stable inclination.

In another co-HTC experiment combining sewage sludge and food waste, Zheng et al. experimented with varying operation conditions and investigated the fuel properties of hydrochars. Co-HTC led to enhanced combustion behavior. The highest energy content is reported as 22.87 MJ/kg for food waste and sludge mix at 230°, (30% sludge – 70% food waste) (Zheng et al. 2019).

Kim et al. hydrothermally carbonized sewage sludge and peat (1:1 ratio) at temperatures varying from 200° to 350° at 60 min, for improving fuel quality. The mixture with a moisture content of 80% showed higher energy content due to H and O reduction in solid products after HTC treatment which is proven to increase the fuel properties of low rank coal (Kim et al. 2017).

Li et al. utilized landfill leachate and activated sludge as liquid product in HTC of paper, food waste and yard waste. The experiments showed that leachate and sludge did not have a significant impact on the final product yield and these sources could be used to sustain the HTC process (Li et al. 2014) which should be further evaluated in relation to leachate and sludge management.

Saetea and Tippayawong produced hydrochars from sewage sludge at 200°, 1–6 h. using oxalic acid as catalyst, increasing the carbon content from 49% to 56–58%. Hydrochars with approximately 23 MJ/kg energy content possessed lignite like qualities. Iodine adsorption capacity of the porous structure was also investigated. LFHTC was tested to have high N and K content (Saetea and Tippayawong 2013) which could be favorable in soil remediation which could only be established with further analyses on material composition including heavy metals. The studies concerning the use of hydrochars as soil ameliorants are reviewed in following sections.

Koottatep et al. processed sludge combined with different catalysts and biomass to produce hydrochars for energy recovery purposes. The influence of catalysts and biomass on fuel characteristics of hydrochars is investigated. An energy content of 28.5 MJ/kg was achieved with optimized HTC parameters of 220°, at 0.5 h retention time with a mixing ratio of 1:0.4:1 for sludge, acetic acid and cassava pulp (Koottatep et al. 2016) making the use of hydrochars a viable option for solid fuel applications.

Gas Production

Parmar and Ross carried out HTC experiments for anaerobic digestates of agricultural waste, sewage sludge, municipal solid waste and vegetable, garden, fruit waste at temperatures 150°, 200° and 250° with 10, 20 and 30% solid loading rates. The investigation of operation parameters on hydrochar quality and biochemical methane production (BMP) revealed that the where agricultural waste produced the highest HHV, sewage sludge digestate generated the highest amount of biogas (Parmar and Ross 2019). Although the application of hydrochars derived from digestate is limited as a solid fuel, its HTC treatment is found efficient for biogas production.

Nyktari et al. investigated the biogas rates from LFHTC of sludge at temperatures 140° – 200° and retention times 30–240 min. and found out that higher biogas production is achieved at lower HTC temperatures (Nyktari et al. 2017).

Villamil et al. treated the liquid fraction of hydrothermal carbonization process (LFHTC) of dewatered sludge with primary sludge by anaerobic digestion (AD). Three different rates of 25, 50 and 75% of LFHTC resulting in an increased methane generation as the loading increased. A chemical oxygen demand (COD) reduction of 86% was achieved. The results suggest that HTC of activated sludge might be a probable addition to wastewater treatment plants (Villamil et al. 2019).

Wang et al. used HTC process to produce hydrochar from high-ash sewage sludge under a wide temperature range of 170° – 350°. The optimum temperature is determined as 230° concerning the combustion characteristics of hydrochars. It is suggested that LFHTC with high levels of COD and TOC can be further treated by AD for biogas production (Wang et al. 2019).

A comparison study on biochars produced from the low temperature pyrolysis and the hydrothermal carbonization of sewage sludge was conducted by Gai et al. (2016). Based on the outcome of the experiments, an integrated approach combining HTC and gasification for syngas production is proposed (Gai et al. 2016a).

Ma et al. (2019) designed an experiment for the co-HTC of sewage sludge and sawdust, coupled with gasification. The results revealed that integration of sawdust to the HTC of sewage sludge increased the calorific value of the produced hydrochar (Ma et al. 2019) while the inorganic compounds suggested applicability for syngas production.

Gai et al. used sewage sludge hydrochars to produce gas rich in hydrogen by using hydrothermal carbonization method followed by steam gasification (2016). It is reported that due to hydrophilic structure of hydrochars from sewage sludge lead to a higher hydrogen yield rendering this raw material efficient for hydrogen rich gas production (Gai et al. 2016b).

De La Rubia et al. (2018) integrated the LFHTC of dewatered sewage sludge with the organic fraction of municipal solid waste (OFMSW) in the AD process. Although the co-digestion did not present beneficial results in means of methane production, a 3:1 ratio (OFMSW:LFHTC) demonstrated similar results to the anaerobic digestion of OFMSW alone, it is suggested as a viable method for LFHTC treatment (De la Rubia et al. 2018).

In a different study concerning the methane production potential of LFHTC of sewage sludge Villamil et al. experimented for the HTC of sewage sludge with a moisture content of 85% at 208°. Methane yields were recorded for AD at 35° (Villamil et al. 2018) and LFHTC is found applicable for methane production under mesophilic conditions.

In a combined approach of HTC and AD, Gaur et al. investigated the energy recovery potential of sewage sludge. HTC was carried out at 200°, 250° and 300° for 30, 60 and 120 min. coupled with AD. Combustion characteristics of hydrochar and BMP of LFHTC were calculated. Energy recovery rates of hydrochar and methane varied between 54–83%, depending on operation parameters (Gaur et al. 2020).

Soil Remediation

Ren et al. (2017) investigated the effects of sewage sludge hydrochar for soil remediation purposes, focusing on the cadmium uptake of plants. Hydrochars produced under different operation parameters used for soil remediation purposes are observed to increase the microbial activity and lead to Cd stability in the soil, decreasing the Cd uptake of plants (Ren et al. 2017).

A combined approach of HTC and pyrolysis of sewage sludge is employed by Paneque et al. to produce hydrochars for soil remediation purposes. Moreover, N and P recovery from sewage sludge by this method is found suitable since other disposal methods do not enable such recovery (Paneque et al. 2017).

With the same methodical approach Paneque et al. further investigated the effects of sewage sludge derived hydrochar and pyrochar as a fertilizer for *Lolium perenne*. Pyrolysis process is performed at 600°, whereas HTC is carried out at two different temperatures 200° and 260°. As a result of the experiment, hydrochars produced at lower temperature rates presented a higher yield in plant growth (Paneque et al. 2019), while establishing a more cost effective structure, proving to be relatively environmentally friendly due to lower gas emissions.

Tasca et al. hydrochars produced from the HTC of sewage sludge is assessed to be an option for use as a solid fuel or for soil remediation purposes. It is further stated that micro pollutants and metal content of hydrochars need to be examined for the latter (Tasca et al. 2019). Further research is required to scrutinize the commercialization of HTC technology.

For investigating the soil amendment properties of sewage sludge derived hydrochars specifically on the growing potential of rice, beans and radish, Melo et al. conducted a series of experiments which revealed an acceptable application range of hydrochars for agricultural purposes (Melo et al. 2019). They also emphasized the need for a standardized HTC treatment for specific plant and soil types.

Breulmann et al. explored the elimination potential of phytotoxic compounds of sewage sludge derived hydrochars for plant growth by washing procedure. In the experiments primary sludge, unstabilized and stabilized activated sludge were used. HTC was conducted at 180° – 200° for 4–8 h. Stabilized activated sludge was treated by pyrolysis, where other materials were subjected to HTC. The washing procedure applied to hydrochars is reported to decrease their soil amelioration value due to nutrient loss (Breulmann et al. 2018). Although the hydrochars had a negative effect on plant growth, it is stated that there is room for optimization of treatments for hydrochars to be used for soil amendment purposes.

Chu et al. (2020) investigated the use of hydrochar produced from sewage sludge in regards to nitrogen caption of paddy soil and rice yields. Medium solution applied in HTC consisting of magnesium citrate and H_2SO_4 resulted in highest N uptake of rice and NH_3-N retention of soil (Chu et al. 2020), promoting hydrochar adsorbents for rice production.

Melo et al. examined the effects of hydrochar produced from sewage sludge on earthworms. Hydrochars produced at 190° were then digested using microwave method for trace metal analyses. Hydrochars demonstrated potential as an alternative to commercial fertilizers without an acute toxic effect on earthworms (Melo et al. 2017). However, it is emphasized that further research needs to be carried out for determining the long-term behavior of hydrochar applications for soil amendment.

Zhang et al. used pinewood saw dust with sewage sludge to investigate the hydrochar characteristics, reporting that the co-HTC of these two elements at certain mixing ratios lead to an increase in hydrochar yields, carbon content and the adsorbent structure of the hydrochar (Zhang et al. 2017). The decrease in phosphorus mobility is further noted which is also promising for soil amendment purposes.

Villamil et al. investigated the application potential of hydrothermally carbonized activated sludge as activated carbon. HTC was performed at 140° – 220°, at 0.5–4 h. Chemical activation of hydrochars were carried out with K_2CO_3, KOH, $FeCl_3$ and $ZnCl_2$. KOH and K_2CO_3 were proven to be the better alternative due to the measured adsorption capacity of the produced carbons (Villamil et al. 2020a). Hydrochars produced are also evaluated for soil remediation applications and found to comply with the requirements.

Nutrient Recovery

Xu et al. employed HTC method for phosphorus (P) recovery from sewage sludge and determined the effects of system parameters on P behavior during the process. As HTC enabled the phosphorus transfer to the solid product, addition of CaO assured the solidification of remaining phosphorus concentration in LFHTC (Xu et al. 2018).

In a different to approach to phosphorus recovery from sewage sludge Becker et al. proposed a combined system of HTC, acid leaching, followed by phosphate and nitrogen precipitation. Hydrochar is produced from a digested sewage sludge rich in aluminum and iron salts. LFHTC is used as an ammonium source in the acid leaching process. Nitric acid addition in HTC enhances the carbonization while increasing phosphate recovery. An 80% recovery rate was reported (Becker et al. 2019).

Shen and Zhang (2020) utilized sewage sludge for preparing N-doped porous carbons. Sludge protein was obtained by the HTC of sewage sludge and hydrochars were produced at 240° and 260° with cellulose and the liquid fraction of sludge protein. Hydrochars were then chemically activated by KOH. The produced porous carbons were tested for toluene adsorption. Due to N-doping via sludge protein and the surface area of the porous carbon, the adsorption capacity was increased. HTC treatment is also found to increase adsorbent yields (Shen and Zhang 2020). Moreover, thermal desorption of spent porous carbons is promising in means of regeneration and therefore sustainability of the process.

In another study by Marin-Batista et al. HTC of digested sewage sludge was used for phosphorus removal. HTC carried out 180° – 240° produced low-rank hydrochars, whereas acid leaching performed at 180 with CaO not only increased the hydrochar yields up to 20.5–23.1 MJ/kg, but also enhanced phosphorus sorption (Marin-Batista et al. 2020).

Volpe et al. subjected the hydrochars produced via HTC (190° and 200°, 1 and 3 h) of digested sewage sludge to sequential acid leaching with HCl and phosphate precipitation of the leachate with NaOH for phosphorus recovery. A P recovery rate of 71 wt.% was reported. While the energy content of 22–24 MJ/kg for hydrochars (Volpe et al. 2020) indicates suitability for solid fuel applications, the reduction of heavy metals suggests compliance with soil amendment purposes.

Water Treatment/ Pollutants Removal

Shen et al. (2018) adapted a combined approach with HTC and gasification for nitrogen (N) removal from sewage sludge. HTC was performed at temperatures between 160° – 240° and optimized due to observed N transformation in line with operation costs at temperatures under 200°. 50% of N was removed and the hydrochars obtained were used for the co-gasification process with waste leaves containing Ca. The combined system was reported to reduce the overall NH3 emissions by 57% (Shen et al. 2018).

Another study towards the removal of nitrogen from sewage sludge is conducted by Liu et al. where hydrochar produced via HTC are subjected to pyrolysis. Total nitrogen emissions were less than half of that from the pyrolysis of sewage sludge (Liu et al. 2017a), promoting HTC as a beneficial method for emissions reduction.

Vom Eyser et al. determined the pharmaceutical micro pollutants in hydrochars produced from the hydrothermal carbonization of sewage sludge. Reduced concentrations of pharmaceutical components suggests the potential of HTC treatment (Vom Eyser et al. 2015).

Vom Eyser et al. further performed the HTC of sewage sludge from WWTPs at 210° for 4 h to investigate the pharmaceutical micro pollutants in hydrochars produced. HTC treatment is found efficient to remove or decrease the concentrations of most pharmaceutical residues (vom Eyser et al. 2015). Unlike other micro pollutants investigated within the context of the research, phenazone concentrations were increased after HTC. It is evident that the further research is required to fully comprehend the effects of HTC on pharmaceutical loads.

Belete et al. focused on the effect of LFHTC sludge on microalgal growth of *Coelastrella sp.* and *Chlorella sp.*, declaring no inhibition on the growth rates of microalgeas. A reduction rate of 90% for N and P is reported (Belete et al. 2019), enabling the reuse or release of LFHTC to the environment.

Luo et al. proposed a method for Pb (II) removal from waters via HTC. Mg and Al cations used for sludge conditioning enabled the formation of hydrochars and layered double hydroxides at the same time. The adsorption capacity of these composites towards Pb(II) show that MgAl-layered double hydroxide composites with sludge derived hydrochars are efficient for a combined application for Pb(II) removal (Luo et al. 2020).

Through the chemical activation of hydrothermally treated sewage sludge with KOH, Tasca et al. analyzed the adsorption capacity of produced activated carbons for the herbicide terbuthylazine. With a plausible adsorption capacity, 86% of the pollutant was removed by the sludge based activated carbons (Tasca et al. 2019).

Chung et al. used sewage sludge based hydrochars to prepare adsorbents for the removal of pathogens from wastewaters. Hydrochars obtained at 210°, 5 h were powdered and washed before being centrifuged. Column experiments through sand filtration revealed the adsorption efficiency of hydrochars for rotavirus and adenovirus. Removal rates of both viruses were observed to increase under all testing parameters with hydrochar adsorbents (Chung et al. 2015).

In a similarly structured experiment Chung et al. (2017) used hydrochars derived from sewage sludge for the removal of Escherichia coli from wastewater. Sand columns were fed with raw and chemically activated hydrochar. A removal efficiency of 90% was achieved with KOH activated hydrochars, where columns fed solely with sand demonstrated only a 55% efficiency rate for the same pathogen (Chung et al. 2017).

Ferrentino et al. prepared three hydrochars from sewage sludge for examining their dye removal efficiency from wastewaters. HTC was carried out at 190°, 220° and 250° for 3 h. Raw hydrochars and KOH activated hydrochars were tested for removing methylene blue. Although both materials were effective at low MB concentrations, only KOH activated hydrochars were useful for high concentrations. Around 95% removal efficiency was achieved with chemically activated hydrochars (Ferrentino et al. 2020). While demonstrating the influence of chemical pre-treatment on adsorbent quality, the research indicates HTC process potential for dye removal from wastewaters.

Chen et al. proposed a new method for organic pollutant removal by “visible light molecular oxygen activation” of hydrothermally carbonized sewage sludge and oxalate. Sewage sludge derived hydrochars were proven to be effective catalysts for the degradation of organic pollutants (Chen et al. 2018). Activation of FeIII-oxalates were enhanced by sewage sludge composition under visible light, proposes a method for sludge and water treatment.

Niinipuu et al. (2020) treated different wet material samples by HTC to observe surface properties in relation to their adsorbent characteristics, specifically for methylene blue removal efficiency. Sewage sludge, biosludge, fiber sludge and horse manure were treated at 180°, 220° and 260°. Adsorption capacities of hydrochars decreasing with increasing temperatures regardless of the enhanced surface areas of hydrochars (Niinipuu et al. 2020) drew attention to the critical importance of operation condition optimization in relation to feeding material properties.

Tasca et al. produced sewage sludge based hydrochars from HTC process at relatively high temperatures of 550° – 750° for investigating their potential as an adsorbent for terbuthylazine removal. The chemically activated carbons using KOH performed at a removal rate up to 64%. Increase in chemical activator and temperature increased the adsorption capacity, however resulted in lower solid yields (Tasca et al. 2020). KOH activation and optimization of operation conditions are reported to reduce the ash content.

Kirschhöfer et al. (2016) produced sewage sludge based ACs via HTC for the removal of organic micro pollutants from wastewaters. Five different forms of xenobiotics (sulfamethoxazole, diclofenac and bezafibrate, carbamazepine and atrazine) were removed with an efficiency of 50–80% from WWTP effluent (Kirschhöfer et al. 2016), which indicates potential for optimization specific to micro pollutants in question.

Alatalo et al. investigated the adsorption behavior for Pb, Cr, Cr, and As of municipal sludge and pulp and paper industry sludge subjected to hydrothermal carbonization. The adsorption capacity for both hydrochars is maximized for Pb, where municipal sludge derived hydrochar is reported to have a computable effect on Cr and As (Alatalo et al. 2013). It is found that the end product is promising for metal removal from water.

Wei et al. (2018) investigated the performance of hydrochars obtained from the HTC of municipal sewage sludge (at 180°, 4 h) as an adsorbent for crystal violet. Adsorption kinetics and thermal behavior were observed in relation with the adsorbent dosage, reaction time and pH (Wei et al. 2018).

Spataru et al. used KOH activation to examine the adsorption properties of hydrochar from sewage sludge specifically for orthophosphate and copper removal from wastewaters. KOH activated hydrochar had a higher Cu adsorption capacity than the hydrochar (Spataru et al. 2016). Despite having a lower adsorption capacity than other adsorbents, the chemically activated hydrochar derived from sewage sludge is still considered a valid alternative due to its organic content and availability and in means of sludge disposal.

Liu et al. produced heteroatoms doped porous carbons (HAPCs) using sewage sludge as raw material for hydrothermal carbonization. With the chemical activation of obtained hydrochars, the adsorbent is found to be a both technically and economically feasible material for wastewater filtration (Liu et al. 2017b).

For the hydrothermal treatment of sewage sludge with high moisture content of 88.86% Khoshbouy et al. prepared hydrochars by HTC at 170°, 200°, 230° and 260°. Hydrochars were physically and chemically activated by CO_2 and KOH. KOH activation at a relatively lower temperature of 700° led to a considerably larger surface area than that of CO_2 activated hydrochars. Hydrochars produced at 260° having the best surface area and porosity values were used to remove basic dye methylene blue from an aqueous solution. The adsorption performance of hydrochars was determined to be higher than commercial activated carbons (Khoshbouy et al. 2019). Both the elimination of the drying process and superiority over other activated carbons is promising in means of adsorbent applications of sludge driven hydrochars.

Energy Storage - Supercapacitor Production From Hydrochars

Feng et al. (2016) used HTC process for the production of hierarchical structured carbon from bagasse, employing sewage sludge as an additive. HTC process is combined with KOH activation for obtaining carbon based electrodes. Capacitance is measured as 70.8% at current density of 50 A/g, with a high duration stability of 15.000 cycles at 10 A/g (Feng et al. 2016).

Xu et al. (2020) developed a porous N-doped carbon material through LFHTC carried out with sewage sludge. N-hydrochars were produced by the co-HTC of LFHTC and cellulose. Selected N-hydrochars were activated by KOH for obtaining N-doped carbons. N-carbons were used for the preparation of supercapacitor electrodes. A capacitance rate of 76% at current density of 20 A/g was achieved and the capacitance could be sustained for 10.000 cycles for the supercapacitors (Xu et al. 2020).

Brief information on above reviewed research studies are compiled in Table.1 with reference to feedstock, method, end product and related application field.

Results And Discussion

A networking mapping is generated for all reviewed research material evaluated above, in order to visualize the correlation between relevant research subjects and their density in the research field, with respect to selected keywords representing conducted studies. The findings are shown in Fig. 1, demonstrating the information derived from the software tool VOSviewer, developed for bibliometric analysis by processing big data (van Eck and Waltman 2010).

Focusing on HTC process, the corresponding research areas for sewage sludge valorization are diversified. As categorized above, HTC products of sewage sludge are applicable in fields concerning energy production, environmental and agricultural remediation and energy storage.

Utilization of solid fuels produced from the hydrothermal carbonization of sewage sludge is a highly recognized option among other applications. Literature data for solid fuel combustion focuses on the energy content, chemical structure and process optimization for energy recovery.

Research on gas production via HTC of sewage sludge mostly focuses on biogas production coupled with AD. Utilization of sewage sludge is a suitable method for sludge disposal and clean gas production.

Another available application for sludge derived hydrochar is soil remediation, showing high potential as substitute for commercial fertilizers. Biowaste based fertilizers are a heavily investigated topic, subject to further development. The key component here is the standardization and speciation of product characterization with respect to soil, seed and plant properties.

In pursuit of a circular economy, the holistic approach introducing water treatment and pollutant removal using sewage sludge as the feeding component is quite attractive. Similarly, nutrient recovery proposes enhancement of the overall production cycle.

Due to the porous structure of carbonaceous material activated carbon production for realizing the adsorbent characteristics of this product via chemical activation has gained importance. The use of bio-based sources in supercapacitor production for energy storage is consistently investigated; however integration of sewage sludge derived material is in fact limited. Energy storage from sludge based products is considered a relatively open field for further research, promising for generating high value products and environmental sustainability.

Conclusion

In this review currently available research publications on the hydrothermal carbonization of sewage sludge are assessed and summarized in relation to the investigated feedstock, method, end product and related application field. Applications for the sustainable utilization of sewage sludge are evaluated under six different categories as solid fuel production, gas production, soil remediation, nutrient recovery, water treatment and energy storage.

HTC is a relatively new technology for sludge valorization with a potential for implementation in several applications, enabling a diversified utilization of process products. In light of realized work, it is evident that further research concerning operation parameters, system kinetics, conversion mechanism and compositional structure pertaining to the specific use of products for desired purposes is required for HTC of sewage sludge to be facilitated at a larger scale. For this valorization approach to be fully recognized as a functional tool for sustaining a circular economy, the research should be extended as to cover the environmental and economic implications for each specific application area.

Declarations

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Table

Table 1 is available in the Supplementary Files

Figures

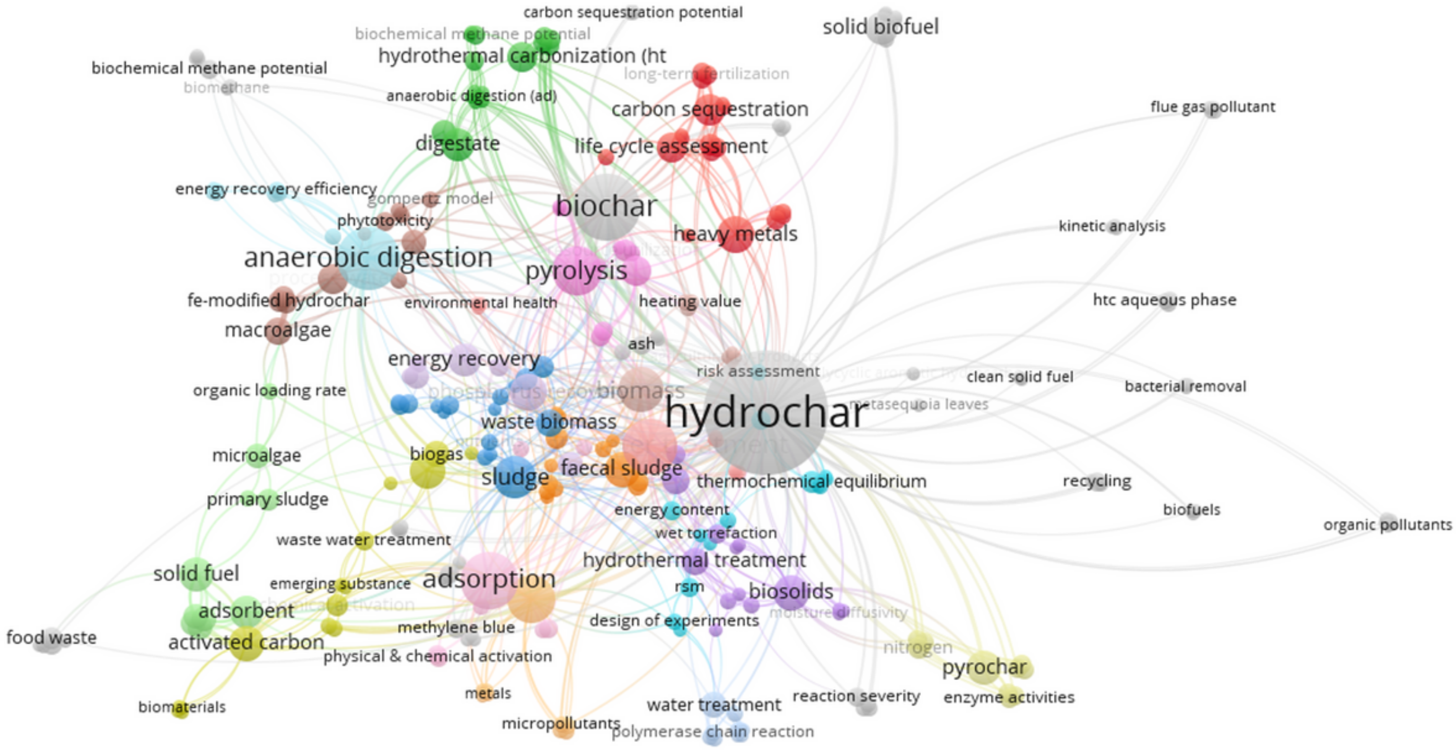

Figure 1

VOSviewer mapping of reviewed research regarding the hydrothermal carbonisation of sewage sludge

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