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A SUCCINCT REVIEW OF CATALYST DOLOMITE ANALYSIS FOR BIOMASS-MSW PYROLYSIS/GASIFICATION[*]

**Obid Tursunov[1,2,3][**], Khairuddin Md Isa[2], Nurislom Abduganiev[1],
Bakhadir Mirzaev[4], Dilshod Kodirov[1], Abdusaid Isakov[5], Sergii A. Sergiienko[6]**

[1]Tashkent Institute of Irrigation and Agricultural Mechanization Engineers, Department of Power Supply and Renewable Energy Sources, 100000 Tashkent, Uzbekistan
[2]Universiti Malaysia Perlis, School of Environmental Engineering, Arau 02600, Perlis, Malaysia
[3]AGH University of Science and Technology, Faculty of Mining Surveying and Environmental Engineering, Team of Environmental Engineering and Biotechnology, 30-059, Krakow, Poland
[4]Tashkent Institute of Irrigation and Agricultural Mechanization Engineers, 100000 Tashkent, Uzbekistan
[5]Tashkent Institute of Irrigation and Agricultural Mechanization Engineers, The Faculty of Electrification and Automation of Agriculture and Water Economy, 100000 Tashkent, Uzbekistan
[6]University of Aveiro, CICECO – Aveiro Institute of Materials, Department of Materials and Ceramics Engineering, 3810-193 Aveiro, Portugal

Abstract

MSW pyrolysis and gasification are possible alternatives to the direct use of fossil fuel energy. MSW, a CO_2 neutral source of renewable fuel, can contribute to the demand for heat, electricity and synthesis gas. However, there are inefficiencies in the technology, which at present render MSW pyrolysis (gasification) economically unviable. The presence of condensable organic compounds and methane in the product gas renders the gas unsuitable for specific applications. Elimination of the condensable organic compounds and methane by a suitably cheap technology will enhance the economic viability of MSW pyrolysis and gasification process. This paper contains a succinct review of the dolomite as the catalyst, which has been evaluated for the elimination of these hydrocarbons/tars and for more qualitative production of gaseous yields from biomass-MSW.

Keywords: catalyst, gasification, MSW, pyrolysis, syngas, tar

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[**] Corresponding author: obidtursunov@gmail.com

1. Introduction

Since few decades, a number of efforts have been initiated to decrease a reliance on fossil fuels, the burning of which leads to release of a significant amount of carbon emissions causing the global issues such as global warming and climate change. Likewise, there is a shift from non-renewable energy sources to bio-energy due to continuous depletion of fossil fuels. Moreover, since bio-fuels are derived from biomass sources, they essentially decrease emissions of harmful gases such as SOx and NOx (Tursunov et al., 2018). Examples of commonly used biomass include plant matter such as forest residues (branches, dead trees, and tree stumps), yard clippings, various energy efficient (high calorific) and fast growing plants, woodchips and municipal solid waste (MSW).

In addition, rapid urbanization and industrial growth, as well as rapid population increase, have led to serious waste management problems in not only developing countries but also in developed countries. In addition, the production of MSW has quickly increased worldwide owing to the fast urbanization and economic development (Cheng and Hu, 2010). MSW contains recyclable materials and energy-containing components, and it is also a resource for misplacement (Iaquaniello et al., 2018). Municipal solid waste disposal into landfills has become a global concern with the increasing tension of energy and resources (He et al., 2009).

On the other hand, rising environmental issues (i.e. greenhouse gases emission and air pollution, etc.) and the energy insecurity resulted from natural resources, like fossil fuels, are driving societies to search for renewable, clean, sustainable and cost-efficient energy supply (Bhuiya et al., 2016; Fulton et al., 2015). Prominent renewable energy portfolios contain biomass, solar energy, hydropower, and wind energy. Unlike other renewable energy sources and among these options, biomass, as an abundantly green and carbon-neutral energy source, can not only be used as a raw material in biomass-powered plants, but it also enjoys a high potential for producing value-added bio-yields (Nikoo and Mahimpey, 2008; Sims et al., 2006; Umeki et al., 2009). MSW as one of the dominant biomass sources, its treatment has led many research-scholars to pay attention to the use of MSW as an energy source (Hong et al., 2017). The high carbon content and volatiles in MSW has transformed their pyrolysis or gasification into an attractive option to produce high caloric (energetic) value syngas with low tar content.

Remarkable progress has been achieved in recent years in the design of gasifiers. However, gas cleaning is still the bottleneck in advanced gas utilization that limits the deployment of the use of MSW for electricity generation (Kaltschmitt and Rosch, 1998). The continual build-up of condensable organic compounds (often referred to as tars) present in the product gas can cause blockages and corrosion and also reduce overall efficiency. In addition, the presence of impurities (such as methane) can affect the end of syngas usage and the techniques involved in the removal of the impurities in such processes are costly. Nitrogen and Sulphur are present in many of the by-products and the corresponding oxides are produced during combustion of the fuel gas; these oxides (NO_x and SO_x) can have a negative environmental impact (Chen et al., 2014; Zhang et al., 2015).

Since the mid-1980s, interest has grown on the subject of catalysis for MSW pyrolysis and gasification. The advances in this area have been driven by the need to produce a tar-free product gas from the pyrolysis and gasification of MSW since the removal of tars and the reduction of the methane content increases the economic viability of the MSW pyrolysis/gasification process. The literature in this area ranges from papers on bench-scale reactors to those on the use of plant-scale gasifiers (Aznar et al., 1993; Chen et al., 2014; Lindman, 1981; Ramos et al., 2018; Shah, 2014).

Research on catalysts for use in the process is often carried out specifically in relation to gasifier design or MSW feed type. However, the criteria for the catalyst are fundamentally the same and maybe summarized as follows:

- the catalysts must be effective in the removal of tars.
- if the desired product is syngas, the catalysts must be capable of reforming methane.
- the catalysts should provide a suitable syngas ratio for the intended process.
- the catalysts should be resistant to deactivation as a result of carbon fouling and sintering.
- the catalysts should be easily regenerated.
- the catalysts should be strong.
- the catalysts should be inexpensive**.**

Thus, this review study aims to provide comprehensive information on dolomite as a catalyst for the transformation of MSW into gaseous yields. Times New Roman 10pt single

2. Catalyst dolomite for MSW pyrolysis/gasification

Dolomite, a magnesium ore with the general formula $MgCO_3 \cdot CaCO_3$ is used in the Pidgeon process for the manufacture of magnesium by thermal reduction. The use of dolomite as a catalyst in MSW pyrolysis (gasification) has attracted much attention since it is a cheap disposable catalyst that can significantly reduce the tar content of the product gas from a gasifier. It may be used as a primary catalyst, dry-mixed with the MSW or, more commonly, in a downstream reactor, in which case it is often referred to as a guard bed.

The chemical composition of dolomite varies from source to source but it generally contains 30 wt.% CaO, 21 wt.% MgO and 45 wt.% CO_2; it also contains the trace minerals SiO_2, Fe_2O_3, and Al_2O_3. The surface areas of the various types also differ, as do the pore sizes and distributions. Orío et al. (1997) investigated four different dolomites (from Norte, Chilches, Malaga, and Sevilla) for oxygen/steam pyrolysis (gasification) of wood in a downstream catalytic reactor. The main chemical difference between the various samples was the Fe_2O_3 content: the Malaga and Sevilla dolomites had low levels of Fe_2O_3 compared to those from Norte and Chilches. These samples were tested as catalysts at varying steam carbon ratios and temperatures ranging from 805 to 875°C.

Tar conversion was of the order of 95% for the Norte dolomite and the lowest conversion of 77% was found for the Sevilla dolomite. The gas yields were increased by the catalyst for all of the dolomites. The order of activity was: Norte>Chilches>Malaga>Sevilla. Interestingly, the surface areas of the Norte and Chilches dolomites were lower than those of the Malaga and Sevilla materials. The higher activity of the Norte and Chilches dolomites may be accounted for by their higher Fe_2O_3 contents and also by their larger pore diameters. The increase in gas yield was 10–20 vol.%, resulting in an increase of 15% in the "lower heating value" (LHV) of the gas. The hydrogen content of the gas increased by 4 vol.%, while the content of CO, CO_2, and CH_4 was relatively unchanged. Perez et al. (1997) and Aznar et al. (1997) also investigated the use of Malaga dolomite for steam/oxygen pyrolysis (gasification). They reported that the H_2 content of the flue gas increased by 7 vol.%, while the CO content decreased by 7 vol.%. This effect was due to a greater contribution of the water-gas shift reaction as a result of a high steam content and high temperature (Perez et al., 1997).

In an earlier paper (Aznar et al., 1997) under different pyrolysis (gasification) conditions, in which the CO_2 content varied slightly from −2 to +6 vol.%, an overall decrease in the CO_2 was reported, indicating that dry reforming also occurred. The methane and steam contents also decreased by 0.8–2.0 and 3–8 vol.%, respectively, as a result of the water–gas shift reaction (Perez et al., 1997). The LHV of the gas increased by 10% to 21% while the gas yield increased by 0.15–0.4 m^3 (Aznar et al., 1997). A tar conversion of 96% was

obtained at a temperature of 840°C (Baker et al., 1996; Richardson and Gray, 1997). Ekström et al. (1996) also achieved almost 100% conversion of tar at 700–800 °C using Malaga dolomite under steam reforming conditions. However, they also observed a marked increase in CH_4 and C_2H_4 at lower temperatures and showed that calcined dolomite was 10 times more active than the uncalcined material (Ekström et al., 1996), in agreement with the results of several authors (Aznar et al., 1996; Baker et al., 1996; Brenes, 2006; Delgado et al., 1997; Ekstrom et al., 1996; Myren et al., 1997; Orio et al., 1997; Richardson and Gray, 1997; Vassilatos et al., 1992). Delgado et al. (1997) also investigated the use of Norte dolomite and compared it with calcite (CaO) and magnesite (MgO) for the steam reforming of MSW tars. They investigated the effects of temperature, contact time and the particle diameter of the catalysts and reported that the tar conversion increased on increasing the temperature of the catalyst bed, complete elimination being observed at 840°C.

The increase in temperature also resulted in an increase in the gas yield. Delgado et al. (1997) reported that the gas composition was unchanged, in contrast to the results of other groups (Aznar et al., 1997; Ekstrom et al., 1996; Perez et al., 1997). An increase in the gas/catalyst contact time gave an increase in the destruction of tars present in the gas, a maximum is being reached at 0.3 kg h/m^{3}n. The increased contact time produced more H_2 and CO_2 as a result of the occurrence of tar conversion reactions and the water–gas shift reaction. An increase in particle size had the same effect as that of increasing the contact time. Delgado et al. (1997) also investigated the effect of calcination on all three catalysts. He reported that the order of activity was dolomite>magnesite>calcite.

Calcined Glanshammar and Sala dolomites were investigated by Vassilatos et al. (1992), who studied the effects of temperature, catalyst contact time and steam/carbon ratio. With the calcined dolomite increasing the temperature, it gave increased gas yields as with the results published by Orio et al. (1997) and by Delgado et al. (1997). The effect on the gas composition was also similar to that described by Orio et al. (1997), this being an increase in the H_2 and CO_2 content. The tar content decreased with increased catalyst loading, for both the Glanshammar and Sala dolomite (Vassilatos et al., 1992).

Vassilatos et al. (1992), Myrén et al. (1997), and Brenes (2006) also investigated the use of Sala dolomite and reported good results for the reforming of tars. The former group reported that the resulting high conversion of the tars with Sala dolomite leaves almost nothing other than naphthalene in the condensed tar phase. Vassilatos et al. (1992) found that the naphthalene production seems to be connected with the steam content and temperature. Several authors (Alden et al., 1992; Brenes, 2006; Cheng et al., 2017; Dalena et al., 2017; Lammers et al., 1997; Myren et al., 1997; Vassilatos et al., 1992) have identified naphthalene as the most abundant condensable product after reforming the tars over dolomite at 800 to 900 °C. This observation highlights the limitations of the use of dolomites as catalysts for the complete elimination of tars from the product gas. Aldén et al. (1992), Lammers and Beenakers (1996), Cheng et al. (2017) and Dalena et al. (2017) investigated the catalytic reforming of naphthalene over dolomite. The former group (Alden et al., 1992) reported that the degree of conversion of naphthalene when passed over calcined dolomite at 800 °C varied with the composition of the carrier gas.

A conversion of 96% was achieved using a carrier gas composition of 15% CO2 (balance N2), while only 79% conversion was achieved with 18% H2O in the gas. The resulting product gas compositions are shown in Table 1. The degree of conversion of naphthalene also varied with the amount of water vapor in the carrier gas, reaching a maximum for a water concentration of 5–15 vol.%. Lammers et al. (1997) and others (Cheng et al., 2017; Dalena et al., 2017) investigated steam pyrolysis (gasification) and the effect of air on the reforming of tar over calcined dolomite. The degree of tar conversion at 850°C over the calcined dolomite with steam alone was 72% while 96% conversion was achieved in an air/steam mix at 850 °C. The gas composition exiting the catalytic reactor was only

slightly affected. From micro-reactor experiments, it was found that the addition of air in the catalytic reactor introduced an extra parallel naphthalene decomposition reaction.

Table 1. The product composition (vol. %) when converting naphthalene over calcined dolomite at 800 °C (Alden et al., 1992)

Carrier gas comp, balance N_2	H_2	CO	CO_2	CH_4	Dolomite
14% CO_2	1.6	2.8	16	0.67	No
14% CO_2	2.9	15	4.6	0.13	Yes
18% H_2O	11	4.3	1.9	0.22	Yes

Williams and Horne (1994, 1995), who pyrolyzed biomass in the form of wood in a fluidized bed and upgraded vapors with catalysts in a fixed bed downstream. Furthermore, the details of the materials balance calculation at different reactor temperatures over the range of 750–900 °C were presented in Table 2. The overall material balance had a closure of ~94.40%.

Table 2. Product yields and recovery from catalytic pyrolysis of MSW at different reactor temperature (Williams and Horne, 1994, 1995)

Temperature, (°C)	Gas, (wt%)	Oil, (wt%)	Char, (wt%)	Recovery[a], (wt%)
750	43.42	34.55	18.53	96.50
800	64.84	16.06	16.53	97.43
850	75.63	7.71	15.42	98.76
900	78.87	5.13	14.92	98.92
900[b]	44.07	37.98	15.86	97.91

[a]Recovery (wt%) = Gas (wt%) + Oil (wt%) + Char (wt%), [b]Pyrolysis process.

Therefore, pyrolysis product yields profile confirmed that the catalysts promote depolymerization processes to yield a strong decomposition (low solid yield) or a strong liquid phase cracking (low liquid yield), and hence a higher hydrogen formation since this gas was formed from liquid cracking (Encinar et al., 1997).

In a separate publication, Aldén et al. (1996) reported an investigation of Glanshammar dolomite for the dry reforming of tars from MSW gasifier. With calcined dolomite at 800 °C, they found a 70% reduction of the tar content and a further 10–15% reduction at 900 °C. Tar yields were further decreased when the pressure was increased to 10 bar. However, increasing the pressure also had the effect of raising the partial pressure of CO_2, leading to carbonization of the catalyst. At higher pressure, the function of the catalyst seemed to be more influenced by the partial pressure of the gaseous component than by the temperature. The increased pressure has the effect of increasing the residence time in the catalyst bed and hence of achieving a higher tar conversion.

Simell et al. (1997a) studied a Finnish dolomite catalyst with a relatively high iron content (ca. 1.5 wt.%). The surface area of the calcined dolomite was 8.1 m^2 g^{-1}. The gas atmospheres studied were mixtures of nitrogen as a carrier, with toluene, and H_2, H_2O, CO or CO_2. They showed that the reaction of toluene over dolomite at 900 °C was faster with carbon dioxide than with steam and that dry reforming was inhibited by the presence of steam. Tar decomposition was strongly inhibited by the presence of CO. In a recent publication, Simell et al. (1997b) investigated the mechanism of the catalytic reaction of benzene (as a model compound) over the same Finnish dolomite. Reforming of the benzene was carried out with carbon dioxide. They developed mechanistic models of the Langmuir–

Hinshelwood type for the reforming of benzene and the adsorption of benzene on a single site was identified as the rate-determining step, the CO_2 adsorbing non-dissociative.

Taralas et al., (1993) investigated the steam reforming of cyclohexane over calcined dolomites, testing limestone ($CaCO_3$), dolomitic magnesium oxide [MgO], Sala, Glanshammar and Larsbo dolomites. Using a first-order reaction model, he found that the apparent activation energies were 177.7 kJ mol^{-1} for MgO, 202.8 kJ mol^{-1} for CaO, 153.8 kJ mol^{-1} for Glanshammar dolomite and 168.8 kJ mol^{-1} for Larsbo dolomite.

The catalysts should provide lower activation energy in the transformation of reactants to products. The lower the activation energy, the more efficient the catalysts. Taralas et al. (1993) also reported that the selected dolomites increased the conversion of the cyclohexane at 700 °C without affecting the product gas composition. Tursunov (2014) conducted a comparative study for gas production and analysis from pyrolysis/gasification of MSW with calcined dolomite and zeolite as catalysts, investigated in a downstream fixed-bed reactor between the temperature range of 200-750 ºC. Gaseous yields production from pyrolysis of MSW varied in the range of 49–57 mol%. He claimed that the presence of calcined dolomite influenced significantly the product yields and gas composition in pyrolysis process, and revealed significant catalytic performance on increasing gas yield and decreasing oil yield, and char yield comparing to zeolite and non-catalytic processes (Fig. 1). A higher temperature 600–750 ºC resulted in a higher transformation of MSW into gaseous yields.
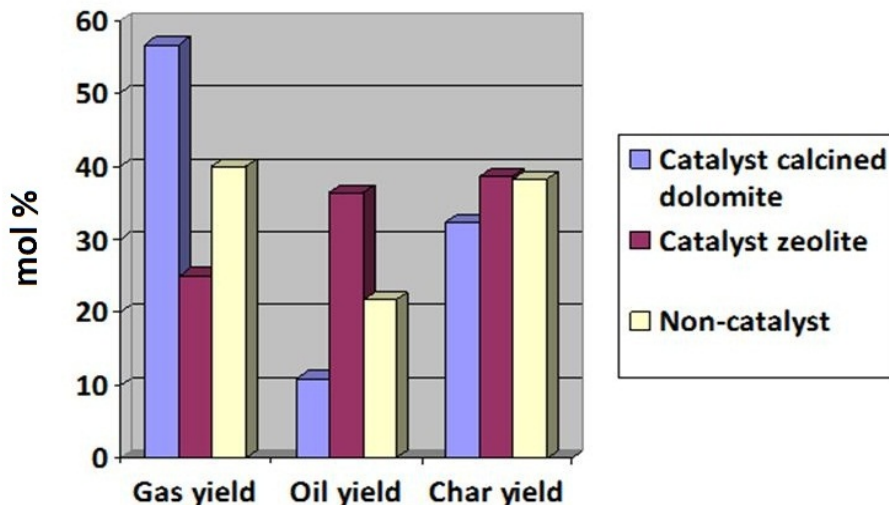

Fig. 1. Comparison of the overall gas, char and oil yield from the catalytic and non-catalytic pyrolysis at 750 ºC (Tursunov, 2014)

Three main groups of catalysts being reported to be successful in removing tars from the producer gas: (1) natural catalysts such as dolomite (subject to further calcinations) (Tursunov, 2014); (2) alkali-based catalysts such as Li, Na, K, Rb, Cs and Fr and (3) metal-based catalyst such as nickel catalysts (Tursunov et al., 2015a; Tursunov et al., 2017). Another study was carried out by Tursunov et al. (2015b) where dolomite and zeolite were used as catalyst for comprehensive analysis of chemical properties (metal oxides). Dolomite was more effective than zeolite as catalyst although is less resistant against attrition and generates more particulates in the product gas. Calcined dolomite demonstrated even better efficiency for increasing hydrogen content as compared to raw dolomite. Moreover, catalysts

as raw dolomite, calcined dolomite (800°C/5h and 950°C/3h) and zeolite have been widely studied (Tursunov, 2014; Tursunov et al., 2015b). It was suggested, that calcined dolomite has an efficiency to eliminate tar from reactors and enhance product yields (e.g. bio-gas, bio-oil, bio-fuele etc.) extraction from the process of pyrolysis or gasification. Regarding to zeolite, it is a catalyst for improving the quantity and quality of bio-fuel yield from pyrolysis technology, but it is not effective catalyst for tar cracking.

One problem reported by Vassilatos et al. (1992) and by other researchers (Corella et al., 1988; Narvaez et al., 1996) was the successive decrease of the mechanical strength of the dolomite with time during all catalytic runs. This decrease was more evident in fluidized-beds. However, once the activity was significantly reduced, the catalysts could be replaced without great expense to the overall process. Deactivation due to carbon deposition was also reported by several scholars (Alden et al., 1992; Brenes, 2006; Corella et al., 1988; Ekstrom et al., 1996; Myren et al., 1997; Narvaez et al., 1996; Simell et al., 1997a; Vassilatos et al., 1992). The relatively high amounts of steam used in pyrolysis (gasification) were effective in maintaining the activities of the catalysts due to the steam reforming of any carbon deposited. Aldén et al. (1992) also showed that the rate of carbon deposition was increased if hydrogen was removed from the gas stream. They proposed that dolomite may catalyze the soot formation and pyrolysis (gasification) reactions. Taralas et al. (1993) reported that the alkaline earth oxides, produced by the calcination of limestone ($CaCO_3$) and dolomites reduce carbon deposition on the catalyst. Lammers et al., (1997), Cheng et al. (2017) and Dalena et al. (2017) showed that when dolomite was used, the addition of secondary air to the catalytic reactor has the important effect of significantly reducing the deactivation rate of the catalyst.

3. Conclusions

Dolomite is a suitable catalyst for the removal of hydrocarbons, which are evolved in the pyrolysis or gasification of MSW. The dolomites increase gas yields at the expense of liquid products. With suitable ratios of MSW feed to oxidant, almost 100% elimination of tars can be achieved. The dolomite catalyst deactivates due to carbon deposition and attrition; however, dolomite is cheap and easily replaced.

The catalyst is most active if calcined and placed downstream of the gasifier in a fluidized-bed at temperatures above 800 °C. The reforming reaction of tars over dolomite occurs at a higher rate with carbon dioxide than steam. Dolomite activity can be directly related to the pore size and distribution. A higher activity is also observed when iron oxide is present in significant amounts.

The dolomites are not active for reforming the methane present in the product gas and hence they are not suitable catalysts if syngas is required. The main function of dolomite is to act as a guard bed for the removal of heavy hydrocarbons prior to the reforming of the lighter hydrocarbons to produce a product gas of syngas quality.

Acknowledgements

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