International Journal of Applied Engineering Research
ISSN 0973-4562 Volume 10, Number 4 (2015) pp. 9245-9256

http://www.ripublication.com

Tar Mitigation In Biomass Gasification Systems Using Catalysts: An Overview

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Abstract:

Biomass gasification is one of the promising technologies for converting biomass to bioenergy**.** It is widely recognized that tar generated along with producer gas presents a significant impediment to the application of biomass gasification system, by getting deposited on surfaces of filters, heat exchangers, and engines, thereby reducing the component performance and invites recurring maintenance problems. Though several tar removal methods are available catalytic tar mitigation method is highly preferred due to its inherent advantages. The purpose of this review is to investigate catalytic destruction of tar formed during biomass gasification process. Furthermore the mode of application of catalyst for tar removal, their effectiveness, and the critical parameters influencing catalytic tar mitigation in biomass gasification systems have been reviewed and discussed in this paper.

Keywords: Biomass, Gasification, Catalyst, Tar mitigation.

1. Introduction

Globally attention is focused towards biomass gasification, as it holds abundant promise towards decentralized power generation[1-3]. Gasification of biomass generates producer gas, comprising of hydrogen (H_2), carbon monoxide (CO), methane (CH_4) along with numerous undesirable elements such as tar, ammonia (NH_3), ash, char and some trace contaminants. The tar formation during the biomass gasification process is one of the major problem which hinders the commercialization of this technology. Tar condenses below its dew point temperature and blocks the downstream process equipments such as engines and turbines, thereby reducing component performance and increases maintenance requirements[4-6]. Moreover, tars are highly carcinogenic, and creates enormous environmental pollution. Tar must be mitigated in biomass gasification systems to make it viable option for energy producers. Enormous studies have been directed towards tar mitigation in producer gas. The methods to remove tar from producer gas can be generally classified into one of the three categories: physical processes, thermal processes and catalytic processes. In physical process, filters or wet scrubbers are employed which just traps the tar, creating waste tar treatment problems and associated environmental problems. The thermal processes raise the temperature of the producer gas around 1000°C to crack the heavy aromatic tar species into hydrogen and carbon monoxide which requires tar cracking system to be constructed of expensive alloys. The catalytic tar cracking can operate at relatively lower temperatures and generates high tar removal efficiency and is recognized as the most efficient method to diminish the tar formation in the producer gas mixture[7-12]. Abu –El-Rub et al. [13] stated that catalytic tar cracking method doesn’t create waste water disposal problems, but on the other hand improves the quality of producer gas. Hence the catalytic tar reduction studies in biomass gasification systems has attracted enormous interest among researchers.

The purpose of this study is to review the catalysts available for tar mitigation, their effectiveness in in-situ and ex-situ modes, and to study the influence of various parameters of catalytic tar mitigation.

2. Catalysts for tar removal in biomass gasification systems

Abu–El-Rub et al. [13] classified the tar cracking catalysts as synthetic and minerals, based on its production methods. The mineral catalysts, dolomite and olivine are cheaper but are less effective in enhancing the quality of producer gas to be employed in applications such as fuel cells, so researchers have focused their attention towards highly active synthetic catalysts. Among the synthetic catalysts the transition metals such as platinum, rhodium, ruthium and nickel posses high tar mitigation capability. Asadullah et al [14] reported the tar conversion capability of the transition metals in the following order Rh>Ru>Pd>Pt>Ni. The Rh, Ru, Pd and Pt catalysts are highly active but are costlier, so research is focused on nickel catalyst which are remarkably active and can be availed at cheaper cost. Furthermore to enhance the performance and stability of the nickel catalysts, various supports and promoters are employed[15-16]. Miyazawa et al [17] experimented with the different supports for nickel based catalysts and reported the tar destruction activity in the following order $Ni/Al_2O_3 > Ni/ZrO_2 > Ni/TiO_2 > Ni/CeO_2 > Ni/MgO$. Chaiprasert et al [18] experimented with Pt, Fe and Co as promoter for nickel catalyst and concluded that Pt metal highly enhanced the performance of Ni catalyst and also suppressed the coke deposition. Coating ceramic elements such as candle filters and monoliths with catalyst layers leads to increased conversion of the tar. However monoliths are more expensive than commercial catalysts and is currently less opted for tar removal [19]. The various tar cracking reactions are detailed below[20-22].

Steam Reforming reaction

$C_nH_m + nH_2O \rightarrow nCO + (n+m/2)H_2$ (1)

Dry reforming reaction

$C_nH_m + n\,CO_2 \rightarrow 2nCO + (m/2)H_2$ (2)

Thermal cracking reaction

$CnHm \rightarrow C^* + CxHy + gas$ (3)

Hydro cracking (or) Hydro reforming of tars

$C_nH_m + n\,H \rightarrow CO + H_2 + CH_4 + \ldots\ldots + Coke$ (4)

Water –Gas Shift reaction

$CO + H_2O \rightarrow CO_2 + H_2$ (5)

where C_nH_m and C_xH_y represent tar and lighter tar respectively.
The above detailed bulk catalyst suffers from disadvantages such as mass transfer resistance, larger time consumption for tar cracking, faster deactivation and creates disposal problems[23]. Of late nano catalysts are employed in tar cracking process to overcome the above stated disadvantages. The nano catalyst possess particle size in the range of 1 - 100 nm. The nano catalyst is expected to perform better than bulk catalysts as they have higher number of active sites per gram[24]. Jianfen Li et al [25-26] conducted experiments using nano Ni-La-Fe/Al_2O_3 and NiO/γ- Al_2O_3 and reported 99% tar removal at a temperature of 800°C. The available nano catalyst based tar reduction studies are very limited and intensive research is to be carried out.

3. Strategy of catalytic tar mitigation

Catalytic tar mitigation can be accomplished by two methods.

(a) One approach involves incorporating or mixing catalyst with the feed biomass to achieve catalytic gasification. This method is one of the primary method in which tar is removed inside the gasifier itself and is termed as in-situ approach.

(b) In the second approach, the producer gas is catalytically treated downstream of the gasifier in a secondary reactor. This method is one of the secondary methods used for tar reduction, whereby the tar is removed outside the gasifier and is termed as ex-situ approach [13].

3.1. Tar cracking effectiveness of in-situ catalysts

In-Situ tar cracking method is cheaper, as it eliminates the need for secondary catalytic converter and guard bed. In-Situ or primary catalysts are added prior to gasification and can be dry mixed with the feed material or can be wet impregnated. Generally these catalysts are non-renewable and consists of cheap disposable material. Arauzo et al. [27] reported phenomenal tar cracking capability of Ni-Mg Aluminates catalyst at 650°C during gasification of poplar feed material, but suffered deactivation due to carbon deposits. Baker et al [28] reported rapid deactivation of G- 90C catalysts due to carbon deposition on catalyst surface. Experiments conducted using nickel based catalysts also reported deactivation of catalyst due to carbon deposition [29]. Furthermore to avoid rapid deactivation of catalysts, the ex-situ approach is more predominantly employed.

3.2. Tar cracking effectiveness of ex-situ catalysts

Corella et al.[30] observed 97-99.5% tar conversion using Ni based catalysts {ICI57-3, UCIC11-9-061, Topsoe R-67, Basf (G1-50)} at 730-850°C, and no deactivation occurred up to 65 hours. Six nickel based catalysts were reported to be active, without being deactivated by coke deposition even after 12 hours[31]. Ex-Situ or secondary catalytic tar reduction has proven to be an effective approach. In addition to the main catalytic reactor the guard bed is employed to protect the tar cracking catalysts. Usage of guard bed comprising of inexpensive catalytic material upstream of metallic catalyst bed has demonstrated to improve the life of metallic catalysts. The inexpensive mineral catalysts such as dolomite used in guard bed converts the heavy tars while the metallic catalysts serves to polish the gas and reduces the tar concentrations to very low levels [12].

4. Influence of parameters on catalyst performance

Several parameters such as catalytic bed temperature, catalyst loading, space velocity, catalyst particle size, Catalyst/feed ratio , calcinations temperature, gas residence time, type of gasifying agents, equivalence ratio, and biomass feeding rate plays an vital role on influencing the effectiveness of the catalysts during tar mitigation and are detailed below.

4.1. Effect of catalytic bed temperature

Nacken et al. [32] reported increment in conversion of benzene and naphthalene model tar compound from 2% to 100% when catalyst bed temperature was increased from 700-900°C when Ni doped catalysts was used. Complete tar cracking of naphthalene model tar compound occurred at 800-900°C when nickel was loaded on a activated candle filter[33]. Bangala et al. [34] reported total napthalene model tar compound conversion at 1023 K using UCI GB-8 catalyst. Wang et al. [35] observed increment in tar conversions from 43-99 % with dolomite and nickel catalysts when catalytic bed temperature was increased from 650-850°C. Naphthalene model tar compound conversion reached 100% at 800°C, using Ni based catalytic filter [36].Tar conversion increased to 98% when catalytic bed temperature was increased from 625-

850°C, when Nickel-A,B, D,E, UC-11-9-62, BASFG1-25S catalysts were used on pine wood chips[37]. Hence it is inferred that increment in catalytic bed temperatures reduces the tar content but increment in energy requirement of the catalytic reactor reduces the overall gas efficiency.

4.2. Effect of catalyst loading

Corella and Caballero et al. [38-40] has demonstrated that nickel loading of >15 wt % was very effective in removing tar in a secondary bed at 1003-1123 K. Tasaka et al. [41] reported 80 % tar conversion in secondary tar cracking using 12 wt % Co/MgO catalyst, in primary method of tar cracking. Wang et al. [42] reported increment in benzene conversion when Ni catalyst loading was increased from 3 to 15 wt %, but tar conversion remained unchanged above 15 wt%, and concluded that optimum Nickel loading is 15 Wt %. Swierzynski et al. [43] observed toluene conversion increment from 12 to 36% when catalyst loading of Nickel-olivine was increased from 1.5 to 5.7 wt %. at 560°C. Different catalyst loadings have been reported to be effective for different catalysts. Hence experiments have to be conducted to validate the exact catalyst loading for a particular catalyst.

4.3. Effect of space velocity

Dou et al [44] reported that tar conversion reached 100% for space velocities of 1000-$^{-}$4500h $^{-1}$ for Y-Zeolites on naphthalene tar compound, but above 7000h^{-1} the activity dramatically decreased, and concluded that catalysts cannot be effectively used at high space velocity. Zhang et al [12] experimented with nickel based catalysts (ICI46-1, Z409 and RZ409) on seed corn fuel in a secondary catalytic reactor and observed that space velocity had little effect on tar cracking and gas composition. It is inferred that these contradictory statements may be valid owing to the different catalytic reactor geometry employed by researchers across the globe.

4.4. Effect of catalyst particle size

Kong et al [45] reported that 7.9 nm Ni/Al2O3 catalyst yielded lower tar than 22.1nm catalyst on toluene model tar compound, and concluded that smaller particle size of catalysts contributed to higher toluene conversion. Aznar et al [46] observed that when particle size was decreased from 2.5 to 0.5 mm lesser tar occurred, when Topsoe R-67-7H, Ni/Al_2O_4 catalysts were used on pine sawdust. Ni/dolomite catalyst size reduction from 5 to 3 mm increased the hydrogen yield[47]. The tar conversion capability of Ni/CeO_2-ZrO_2 catalyst increased from 57.7 to 62.9% when its particle size was decreased from 5 micrometer to 20nm[48]. Hence it is inferred that catalysts of smaller particle size are comparatively more active than bigger sized particles due to its inherent larger surface area.

4.5. Effect of catalyst /feed ratio

Chunfei Wu et al.[49] reported that gas yield increased from 79.1 to 91.9 % wt % when catalyst/feed ratio was increased from 0 to 2 using Ni/CeO_2 /Al_2O_3 on polypropylene feed. Manya et al. [50] reported decrement in tar from 4.75 % of feed to 2.75 % of feed when alumina quantity was increased from 0 to 8 % of feed. Hence it is inferred that increment in catalyst/feed ratio reduces the tar, but further experiments have to be conducted to optimize the ratio for specific catalysts.

4.6. Effect of calcination temperature

Swierczynski et al.[43] experimented on Ni-olivine catalyst on toluene model tar compound with catalysts calcined at 900, 1100 &1400°C and observed that toluene conversion decreased with increasing calcination temperatures. Tar conversion decreased from 99 to 24 % when calcinations temperature increased from 900 to 1400°C. Pansare et al.[51] reported that yield of methane and benzene decreased with increase in calcinations temperature from 400-1000°C, while tungsten zirconia (WZ) was used for cracking of toluene model tar compound. Furusawa et al.[52] reported that CO/MgO catalyst yielded better results on cellulose feed when calcined at 873K.The contradictory statements may be valid owing to the different catalyst configurations employed by different researchers.

4.7. Effect of gas residence time

Maria P. Aznar et al. [46] reported 98% tar conversion for residence time of <0.1 seconds when nickel catalysts (A, B, D, E Uc-11-9-62) was used on pine wood chips. Markus Ising et al.[53] reported total tar conversion for a residence time of 0.3 to 0.4 seconds when monolithic nickel catalyst was used at 900°C on wood chips feed material. Duo Wang et al.[42] reported that 0.3 seconds was the optimum residence time for cracking benzene model tar compound using Ni/Al_2O_3 catalyst. Kazuhiko Tasaka et al.[54] reported 80 % cellulose tar conversion for a residence time of 0.06 seconds using Co/MgO catalysts. Abu-El-Rub et al.[55] reported that 0.3 seconds was the optimum residence time for cracking naphthalene & phenol model tar compound using biomass char as catalyst. Dennis Bangala et al.[34] reported that tar conversion increased when residence time was increased from 0.3 to 0.6 seconds for GB-98 catalyst for cracking naphthalene model tar compound. Hence it is inferred that higher residence time reduces tar and optimum value of residence time depends on the activity of the different catalysts.

4.8. Effect of equivalence ratio (ER)

Tomohisa Miyazawa et al.[56] reported total tar conversion at ER= 0.25-0.35 when Rh/CeO_2/SiO_2 was used for cracking tar from woody biomass. Miguel A. Caballero et al. [57] reported decrement of tar when ER was increased from 0.18 to 0.36 when pine wood chips was cracked using nickel based catalysts (BASF G1-50, ICI46-1, Topsoe R-67). Hence it is inferred that with increase in ER, the gasification process closes on towards combustion process. Hence it is concluded that with increase in ER, the tar content has to decrease, but the calorific value of the producer gas will also get reduced due to conversion of H_2 and CO into H_2O and CO_2.

4.9. Effect of gasifying agents

Lopamudra Devi et al. [58] reported that addition of steam, CO_2 increases naphthalene conversion, whereas addition of hydrogen reduces conversion when olivine was employed. Pattaraporn et al. [59] reported increment in steam (S/C ratio 0.2-1.0) resulted in higher H_2 and decreased CO_2 and CH_4,whereas addition of O_2 showed increment of CO,CO_2 and decrement of H_2,when Ni/Dolomite was used for Coconut shell gasification. Steam reforming reaction gave lower tar conversion than dry reforming reaction. Hence it is inferred that tar conversion and resulting gas composition is a function of gasifying agents.

4.10. Effect of biomass feeding rate

Miyazawa et al [56] reported increment of tar from 0 to 4 % when biomass feeding was increased from 150 to 250 mg/min, when Rh/CeO_2/SiO_2 was used as catalyst. Gervasio et al [60] reported that when naphthalene feeding rate increased to 35 g/Nm^3 tar conversion decreased from 90 to 25 % , when $LaNiO_3$ catalyst was used. Hence it is inferred that tar conversion decreases when loading of biomass is increased.

5. Limitations of catalytic tar mitigation

Spencer and Twig [61] reported that tar cracking catalysts are easily deactivated by coking, poison adsorption, attrition and sintering. Coke formation can be reduced by usage of increased steam and oxygen in gasification systems. Controlling metallic particle size, usage of promoters and additives can also reduce coking. Employing sulfur sorbent materials such as ZnO to adsorb H_2S which forms ZnS can protect the catalysts from sulfur poisoning. Usage of high strength materials such as olivine reduces attrition problem. High cost of catalyst materials is also a major limitation of catalytic tar mitigation process[11].

6. Conclusion

1) Catalytic tar mitigation is a widely accepted technology, which can reduce tar to acceptable levels of usage in end user devices as recommended by international standards.
2) Transition metals such as platinum, ruthium and rhodium. are highly active but costlier. Nickel is used as a substitute for them as it is relatively cheaper and exhibits remarkable tar cracking capability.
3) The mode of application of catalysts can be by in-situ approach or by ex-situ approach.
4) The major limitation of in-situ approach is the shorter lifetime of the catalysts due to deactivation caused by coking, sulfur poisoning, and attrition .So ex-situ approach is preferred rather than in-situ approach
5) Increment in catalytic bed temperatures reduces the tar content but increment in energy requirement of the catalytic reactor reduces the cold gas efficiency.
6) Smaller catalyst particles exhibit higher activity than larger sized particles due to enhanced surface area.
7) Increment in catalyst/feed ratio reduces tar and further research is required to optimize the ratio for specific catalyst.
8) Higher residence time of the gas in catalytic reactor reduces tar.

9) Tar conversion decreases with increment in biomass feeding rate and tar conversion is a function of gasifying agents.
10) Research efforts have to be directed towards synthesis of novel catalysts comprising of newer promoters and supports to nullify deactivation problems. Research has to be conducted on catalytic tar cracking systems using combination of optimized parameters to improve the effectiveness of catalysts. Reduction of the cost of catalytic tar mitigation process provides scope for future research.
11) The nano catalytic tar reduction studies are very limited and needs intense investigation.

Acknowledgements:
The authors acknowledge the financial support of DST, Government of India, New Delhi to carry out this research work under PURSE scheme. One of the authors, Mr. K. Shanmuganandam is thankful to DST, Government of India, New Delhi for the award of DST PURSE fellowship

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