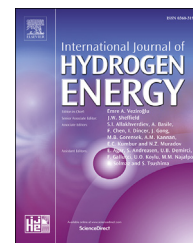


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# Syngas production by catalytic steam gasification of citrus residues

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## ABSTRACT

This study investigate the feasibility to use citrus peel residues to produce hydrogen rich syngas by steam gasification process. Thermodynamic evaluations were performed by simulation modelling thus preliminary experiment was carried out at 1023 K and S/B ratio 1.5 wt/wt. The effect of different types of catalytic materials (minerals and synthetic catalyst) on steam gasification process was investigated in terms of efficiency, hydrogen formation tendency and outlet stream composition.

It is found that concentrations of H<sub>2</sub> and CO of 58.80 mol% and 25.34 mol% can be produced by citrus peels conversion. Moreover, both carbon and hydrogen gasification efficiencies can be enhanced using dolomite, MgO and Ni/Al<sub>2</sub>O<sub>3</sub> in the reactor (from 43.7% to 54.9% and from 34.3% to 54.1% for CGE and H<sub>2</sub>GE, respectively). Dolomite resulted the best catalyst for citrus peels steam gasification under the examined operative condition with CGE and H<sub>2</sub>GE rates of 54.9% and 54.1%, respectively. Ni catalyst resulted to be affected by carbon formation and sintering on time on stream.

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## Introduction

Due to increasing concern about environmental issues such as global warming and depletion of fossil fuels, hydrogen is considered an important energetic vector of the future. In fact, hydrogen can provide for energy in transportation and distributed heat and power generation with no effect on the environment, mainly when it is applied to fuel cells systems [1–5]. However, how to store hydrogen easily and cheaply is still a big problem. In fact, till now, none of the materials and methods for hydrogen storage has been found to meet all requirements for commercial electric vehicle applications [6,7]. Today hydrogen is generated mostly from fossil fuels with a

consequent, release of CO<sub>2</sub> during its production stage, while the use of biomass, as hydrogen tank, represent an attractive alternative to use a renewable source of energy. Hence, studies on biomass thermochemical conversion processes coupled with different reactor configurations have been carried out to serve different purposes, such as developing of fuels processor units [8–10], measuring in the thermography field [11–13], production of biofuels [14–16].

Among biomass typologies agro-industrial residues are obtained in large quantities as a result of industrial processing of fruits and vegetables [17]. The mainly production of citrus fruits juice in Italy is based on oranges and lemons with an annual consume of about 3.8 million tons (Sicily is the main

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Italian fruits producer region with about 87% of the national share). Therefore, every year in Italy, the juice industries produce about 1 million tons of peel waste that represent an environmental problem related to disposal of solid wastes [18]. The conversion of waste materials or industrial byproducts into useful energy can be achieved through thermochemical and biochemical processes [19,20]. Ahmed and Gupta [21], evaluated the performances of both pyrolysis and gasification processes applied to food waste in terms of syngas flow rate, hydrogen flow rate, output power, total syngas yield, total hydrogen yield, total energy yield, and apparent thermal efficiency. They found that gasification was more favorable process for this kind of biomass than pyrolysis, but was noticed that longer time has needed to finish the gasification process. Results also demonstrate that food waste offers a good potential for solid waste thermal treatment with the specific aim of power generation. On the other side, the treatment of orange peels through biological processes resulted inhibited by presence of limonene, which constitutes about 95% of the orange peels oil [22]. Others reported that the bio-oil coming from pyrolysis of citrus wastes has been produced with higher yields than other biomass waste [23,24]. Nevertheless, L. Aguiar et al. [25], revealed that the citrus peel pyrolysis oil can be produced but with low quality due to the high oxygen content, thus it needs a catalytic upgrading prior that it can be used as a fuel or chemical.

On the whole, the specific properties of the processed waste/byproduct/residues significantly affect the conversion performances of thermochemical processes. In particular, some chemical-physical characteristics (i.e. elemental composition, lower heating value, ash content, moisture content, volatile matter content, bulk density, size and contaminants: N, S, Cl, heavy metals, etc.), are so decisive that pre-treatments of the feedstock are often applied before most of prevailing gasification technologies [26,27]. However, gasification has some advantages related to possibility of coupling the operating conditions (in particular, temperature and equivalence ratio) and the features of the specific reactor (fixed bed, fluidized bed, moving grate furnace, plasma reactor) to obtain a syngas suitable for different applications. Furthermore, the gasification process, when conducted using steam as gasifying agent can increase the outlet gas stream in terms of both heating value and hydrogen content [17,26]. The use of catalysts could play a crucial role in increasing the gasification efficiency. In fact, the catalysts promote tar conversion into combustible gas via thermal cracking, hydro-cracking or water gas shift reactions [28,29]. In this context, dolomite and other mineral materials as catalysts has attracted much attention because of their low cost and also can significantly reduce the tars content [30]. At the same time, the Ni-based catalysts are commercially available and they are extensively used for biomass gasification due also to their known activity in naphtha and methane reforming reactions [30].

Although, many papers are present in literature on food waste conversion through thermochemical processes, to the best of our knowledge, there are no works available on the behavior of citrus peel residues working under catalytic steam gasification. Indeed, to date, it is possible to find a research work of the authors developing a non-catalytic air-steam gasification campaign of citrus peel in a fluidized bed reactor [31].

Furthermore, Gadek et al. carried out comparative experiments of air gasification of different biomasses, including citrus peel, in a batch reactor [32]. Accordingly, the aim of this work was to investigate the feasibility to use this type of biomass feedstock to produce hydrogen rich syngas. Preliminary thermodynamic evaluations on citrus peels steam gasification were performed by a simulation modelling; therefore the comparison with experimental data was carried out through a blank no-catalyst test. Successively, attentions were addressed on the effect of different catalytic materials (minerals and synthetic catalyst) on steam gasification process in terms of efficiency, hydrogen formation tendency and outlet stream composition.

## Experimental

### Raw material treatment and characterization

The citrus peels were preventively air dried (383 K for 1 h), shredded and sieved into a grain size range of 25–40 mesh.

Proximate analysis included measurement of moisture, volatile matter, fixed carbon and ash content. The moisture measurement were performed on biomass samples, dried in a convection oven at 383 K until constant weight was recorded. The volatile matter content was determined by measuring weight loss after heating biomass samples to  $1223 \pm 20$  K in an alumina crucible under  $N_2$  atmosphere (ASTM D-2013). The ash content was measured by heating samples (Standard E-1755-01) at  $848 \pm 25$  K for 3 h to constant weight in a muffle furnace and the fixed carbon fraction was calculated by subtracting the percentages of volatile matter, moisture and ash content. Elemental analysis was performed on both biomass samples and bio-char using Thermo Fisher Scientific (Flash EA 1112) CHNS-O Elemental Analyzer. Ultimate and proximate analysis data were reported in Table 1.

Based on the elemental composition and according to literature [14,33,34], High Heating Values (HHV) and Lower Heating Value (LHV) of biomass, were calculated with following equations:

$$\text{HHV(OLS)} = 1.87(C)^2 - 144(C) - 2802(H) + 63.8(C*H) + 129(N) + 20147 \quad (1)$$

$$\text{HHV(PLS)} = 5.22(C)^2 - 319(C) - 1647(H) + 38.6(C*H) + 133(N) + 21028 \quad (2)$$

**Table 1 – Properties of citrus peels.**

Ultimate Analysis (%)						
	C	H	N	S	O <sup>a</sup>	Ash
As received	34.1	5.0	1.0	0.1	32.6	6.9
Dry Basis	42.9	6.3	1.3	0.1	40.8	8.5
Proximate Analysis (%)				HHV	LHV	
Moisture	VM	FC	Ash	(MJ/kg)	(MJ/kg)	
20.0	57.5	15.5	6.9	14.41	13.84	
<sup>a</sup> By difference.						

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