

Cordierite honeycomb monoliths coated with modified forms of ceria for the acid catalyzed synthesis of butylacetate *via* microwave irradiation

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A series of solid acid catalysts like CeO₂, CeO₂-ZrO₂ and SO₄²⁻/CeO₂-ZrO₂ have been coated over monoliths by dip and dry process. Catalysts have been characterized by NH₃-TPD for surface acidity, PXRD for crystallinity, FT-IR analysis for functionality, SEM and TEM for morphology. The catalytic activity of the synthesized materials is trialled by liquid-phase esterification of acetic acid with butanol. The catalytic activity is correlated with acid sites examined by NH₃-TPD method. Optimization of reaction is carried out by altering the molar ratio, temperature, catalyst nature and time of the reaction. A detailed techno-economical analysis of the catalytic material and the procedure followed for the esterification reaction is also studied. In the techno-economical analysis, it is noticed that microwave assisted heating is more advantageous over conventional one. In case of microwave heating, the conversion of BA is 98% with a shorter reaction time of 12 min but in the case of conventional method of heating, it takes almost 6 h to get the above said conversion. It is further noticed that a comparative analysis of honeycomb over powder forms clearly states that, honeycomb forms shows almost 4 fold time increase in the conversion of BA compared to their powder analogs.

Keywords: Acetic acid, Butanol, Modified forms of ceria, Esterification, Cordierite Honeycomb monoliths, Microwave.

Cordierite honeycomb has played a key task as a catalyst carrier in the field of catalysis¹. Honeycomb (HC) monolith supports are uni-body structures and are made up of interconnected repeating cells or channels. Catalysts coated over HCs largely used in automotive applications like selective reduction of NO_x, combustion of volatile organic compounds, ozone abatement in jets etc which involve gas phase reactions². The advantages of catalysts coated on HCs over powder catalysts are highly effective, simple recovery and complete separation of the catalysts from the reaction mass. Applicability of monoliths in the liquid phase as catalytic carriers will be a signifying step in synthetic organic- chemistry³. In the area of catalysis, cerium dioxide (CeO₂) is a multifunctional, promising inorganic compound with wide a variety of technological applications. Incorporation of CeO₂ into ZrO₂ increases the acidity moreover, mixed oxides of ceria-zirconia drastically increases acid-base as well as redox nature.

Esterification is a vital reaction in chemical, pharmaceutical industries. Esters are extensively used in the chemical/pharmaceutical industry as solvents, plasticizers, pesticides, fragrances and flavors. In particular, n-butyl acetate is a colour less organic

solvent having the fruity odour with significant utility both as extractants and dehydrators in the lacquer industry⁴. It can also be used as a solvent in paints and nail polish-removers. Butyl acetate used as an additive to gasoline to prevent engine knocking, artificial leathers, cleaners, and photographic films.

n-Butyl acetate has synthesized by esterification of acetic acid with *n*-butanol by using mineral acids such as H₂SO₄, HCl, etc, as catalysts. But the use of homogeneous catalysts may create problems such as side reaction, corrosion of equipment and difficulty in isolation of product. In addition, the use of homogeneous catalyst involves the neutralization with an alkali, which starts to severe exudation on an industrial scale. In view of these drawbacks, heterogeneous solid acid catalysts are preferred^{5,6}.

A wide variety of catalysts such as ion exchange resin⁷, H-ZSM-5⁸, zeolites-Y⁹, niobic acid¹⁰, sulphated oxides¹¹, heteropoly acid supported clay^{12,13}, and metal ion exchanged montmorillonite¹⁴ have been reported. In all these cases the conventional heating process is adopted. Microwave-assisted organic reactions enhance the chemical reactions from hours to seconds due to its preferential absorption of microwave energy by the polar molecules. Microwave

heating brings quick resulting in reduced reaction time and provides uniform and high efficiency of heating throughout the material. Increased % yield and purity of the product are due to diminished side reactions, because of all these advantages some researchers have switched over from conventional heating to microwave heating in organic synthesis^{15,16}.

Keeping in view of the advantages of microwave heating and use of the catalyst in the honeycomb coated form, in the present work monolith coated with modified forms of ceria was used for the manufacture of butyl acetate from butanol and acetic acid. Manifest applicability of esters, the synthesis of butyl acetate over solid acids such as CeO_2 , $\text{CeO}_2\text{-ZrO}_2$, and $\text{SO}_4^{2-}/\text{CeO}_2\text{-ZrO}_2$ on HC are reported herein. The prepared HC solid acids were characterized for their surface acidity, crystallinity, functionality and morphology. Ceria based solid acids were also prepared in their powder forms. Synthesis of butylacetate *via* esterification of butanol with acetic acid by using solid acids was evaluated in both the forms (powder & HC). To obtain the highest possible yield of butyl acetate, the reaction conditions were optimized by varying the catalyst type, molar ratio, reaction temperature and reaction time. Reusability & reactivation of the catalysts were also conducted.

Experimental Section

Materials and methods

Monoliths with dimensions: height = 1.20 cm, diameter = 2.50 cm and hole = 0.2 cm was supplied by Shreya Ceramics. Zirconyl nitrate, ceric ammonium nitrate, acetic acid, butanol and H_2SO_4 were supplied by M/S SD Fine Chemicals India Ltd.

Preparation and characterisation of catalysts

The metal oxides like alumina or zirconia are wash coated with honeycomb monolith before being coated with the active catalyst. From the literature survey, it is observed that before coating an active catalyst, the bare honeycomb is wash coated with simple metal oxides which can form an adherent coating on the honeycomb surface. When zirconia is wash coated with HC, the surface area increases and also support an HC material for better interaction with the active catalysts compared to other metal oxides¹⁷. For wash coating with zirconia on a bare HC, 1.5 g of zirconyl nitrate was dissolved in 50 mL of deionized water. The resulting solution was coated on HC by dipping and drying in a furnace preheated to 400°C for 10-12 times till ~0.2 g of zirconia was coated on the HC.

(a) Preparation of an HC catalyst by coating with ceria (CeO_2)

A known amount (2 g) of ceric ammonium nitrate was dissolved in 50 mL of deionised water. The solution was coated on a wash coated HC by dipping and drying in a furnace preheated to 400°C for 10–12 times till ~0.2 g of ceria was coated on the honeycomb.

(b) Preparation of HC Catalyst by coating with ceria-zirconia ($\text{CeO}_2\text{-ZrO}_2$)

Calculated amount of ceric ammonium nitrate and zirconyl nitrate were dissolved in 50 mL of deionised water. The resulting solution was coated on a wash coated HC by dipping and drying in a furnace preheated to 400°C for 10–12 times till ~0.2 g of ceria-zirconia was coated on the honeycomb.

(c) Preparation of sulphated ceria-zirconia ($\text{SO}_4^{2-}/\text{CeO}_2\text{-ZrO}_2$)

A known amount of previously prepared hydrated ceria-zirconia solution was taken in a beaker to which 6 M H_2SO_4 (2 mL) was added and the resulting solution was coated on a wash coated HC by dipping and drying in a furnace preheated to 400°C for 10–12 times till ~0.2 g of sulphated-ceria-zirconia was coated on the honeycomb.

Preparation of catalytic materials in powder forms

(a) Preparation of ceria (CeO_2)

A known amount of ceric ammonium nitrate was mixed with a limited amount of deionized water. This mixture was made in to a paste. The resulting paste was dried in an air oven at 120°C for 12 h and calcinated for 5 h at 550°C.

(b) Preparation of ceria-zirconia ($\text{CeO}_2\text{-ZrO}_2$) mixed oxide

Known amount of ceric ammonium nitrate and zirconyl nitrate were mixed with limited amount of deionized water. This mixture was made in to a paste. The resulting mixture was dried in an air oven at 120°C for 12 h and calcined for 5 h at 550°C

(c) Preparation of sulphated ceria-zirconia ($\text{SO}_4^{2-}/\text{CeO}_2\text{-ZrO}_2$)

A known amount of uncalcined $\text{CeO}_2\text{-ZrO}_2$ mixed oxide was taken with a limited amount of deionized water to which 6M H_2SO_4 was added and the resulting mixture was made in to a fine paste. The paste was dried at 120°C for 12 h and calcinated at 550°C for 5 h.

Catalyst characterization

The amorphous or crystalline nature of the catalysts was determined by recording their Powder XRD (PXRD) patterns, recorded on a X-ray

diffractometer (Philips X'pert) using CuK α ($\lambda = 1.5418 \text{ \AA}$). Using graphite monochromator with a 2θ range of 10° with a scanning rate of 2° min^{-1} . The FT-IR spectra of all the catalyst samples were recorded using Nicolet spectrometer instrument by KBr pellet technique with the wave number range $4000\text{--}500 \text{ cm}^{-1}$ for the functionalized samples. The total surface acidity was measured by NH_3 -TPD technique by using Mayura-TPD instrument. In this method, 0.1 g of the catalyst was degassed under N_2 stream for 2 h at 100°C , ammonia (99.99%) gas was injected into the stream until saturation was reached and cooled to 50°C . The system was maintained at 50°C for 30 min and then the catalyst surface was flushed with N_2 for 2 h at 50°C . The catalyst was heated at the rate of 5°C min^{-1} in N_2 to 700°C . SEM and TEM images were recorded on Quanta 200 and JEOL-2000 respectively.

Catalytic activity studies

The catalytic activity of the HC catalysts was verified by liquid-phase esterification of acetic acid with n-butanol. The synthesis of butyl acetate was carried out using acetic acid and n-butanol in the presence of catalysts (powder form & HC) in a microwave reactor (Raga Lab matrix) fitted with a condenser and magnetic stirrer. The reaction was carried out in a specially designed reactor vessel to accommodate the monolithic catalyst (Photo 1). Typically the esterification of acetic acid (10 mmol), butanol (20 mmol) and 0.15 g of HC catalyst were irradiated over the micro oven at a temperature of 80°C at 12 min. The product was analyzed using simple titration with 1N NaOH solution and phenolphthalein as an indicator. The end point of the reaction is colorless to pale pink. The product was also quantitatively analyzed by GC-MS (Varian). The butanol conversion was calculated using the following equation: $X_B = C_B - CB_0 / CB_0$ where X_B is the butanol conversion (%); CB_0 is the initial concentration of the butanol (mol/L) and CB corresponds to the concentration of the butanol (mol/L) at reaction time t (min).



Photo 1 — Microwave reactor with specially designed glass reactor for honey comb reaction.

Results and Discussion

Catalyst characterization

NH_3 -TPD method

The concentration and strength of the acid sites of all the catalyst coated on HCs were verified by the NH_3 -TPD method. The NH_3 -TPD profiles of CeO_2 , SCZ and CZ samples (calcined at 550°C) are depicted in Fig. 1. Desorption peaks at $100\text{--}200^\circ\text{C}$, $300\text{--}400^\circ\text{C}$ and $>600^\circ\text{C}$ were specified as weak, moderate & strong acidic sites.

When sulfate groups adsorbed on the mixed oxide generate strong acidity on its surface, the groups like SO_4^{2-} behave as a Lewis acid or they can generate Lewis acid centers by attracting electrons. Lewis acid sites enhance the Brønsted acid strength of the surface hydroxyl groups. When mixed metal oxides are calcined at a higher temperature, strong acidic active centers are generated on the surface of oxide due to the transformation of ionic to the covalent character. The TPD profile of SCZ shows a strong desorption peak at $300\text{--}400^\circ\text{C}$ and $600\text{--}650^\circ\text{C}$ indicating the existence of strong acidic sites in comparison with ceria as well as ceria-zirconia samples having desorption peaks at 200°C and $300\text{--}400^\circ\text{C}$ respectively^{18,19}. The total surface acidity (TSA) values and acid site distribution of catalysts is given Table 1.

PXRD studies

PXRD outlines of HC catalysts (SCZ, CZ, and C) are enumerated as Fig. 2. Ceria validates a typical

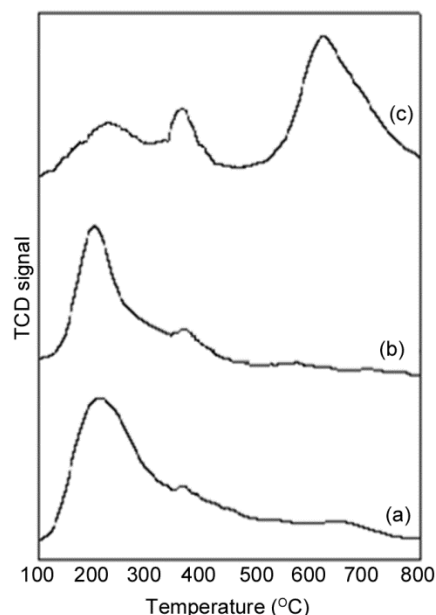


Fig. 1 — NH_3 -TPD profiles of (a) Ceria, (b) CZ and (c) SCZ.

reflection at $\theta = 28.5, 33.4, 47.3$ and 56.2 , with fluorite structure²⁰. The PXRD pattern of ceria-zirconia mixed oxide exhibit characteristic reflections of both fluorite structure of ceria as well as $t\text{-ZrO}_2 = 30.2, 35.1, 50.4$ and 60 ²¹. But in ceria-zirconia, reflections attributable to $m\text{-ZrO}_2$ were absent, thus indicating the tetragonal phase of zirconia stabilized by ceria.

The effect of sulfate ion on ceria-zirconia could be observed with the PXRD pattern of CZ and SCZ are compared and was depicted as Fig. 2. Reflections of $t\text{-ZrO}_2$, the fluorite structure of ceria and two types of sulfates $[\text{Zr}(\text{OH})_2\text{SO}_4]$ and $[\text{Zr}(\text{SO}_4)_2]$ be observed in SCZ²².

FT-IR analysis

FT-IR spectra of C, CZ, and SCZ(HC) catalysts are shown in Fig. 3. A peak at 1400 cm^{-1} is because stretching vibration of Ce-OH. Stretching as well as bending vibrations of OH group of H_2O molecules in the sample are observed²³ at 1600 cm^{-1} and 3500 cm^{-1} . IR-spectra of ceria, ceria-zirconia and sulfated-ceria-zirconia samples exhibit a peak at 3500 cm^{-1} caused by OH group of the catalyst adsorbed on its surface. Similarly, all the above samples have a peak at 1600 cm^{-1} as a result of the deformation mode of OH group present on its surface. Sulfated samples show a peak at $1200\text{-}1400\text{ cm}^{-1}$ indicates the asymmetric stretching frequency (S=O), which represents a typical peak of SCZ²⁴.

SEM analysis

SEM images of HC coated with ceria, ceria-zirconia, and sulfated-ceria-zirconia as well as uncoated HC are given in Fig. 4. From the SEM images it was observed that the surfaces of the honeycomb are uniformly coated by the active catalyst. No distinct features could be observed either in the SEM images of ceria or ceria-zirconia, which signifies the establishment of the homogeneous solution in solid phase between ceria and zirconia. SEM images of sulfated-ceria-zirconia loaded on HC exhibits characteristic feature apart from ceria or ceria-zirconia, which may be due to sulfation and one, can see clear agglomeration in the form of distinct cracks.

TEM analysis

TEM image of SCZ catalyst coated on HC is shown in Fig. 5. TEM image shows that mixed oxides consist of stacked nano-domains with typical sizes $\sim 5\text{-}10\text{ nm}$, indicating the establishment of a homogeneous solution in solid phase between ceria and zirconia.

Table 1 — TSA and acid site distribution catalysts used in the present study.

Catalyst	Acidic site distribution (mmol /g)			TSA
	Weak	Moderate	Strong	
CeO ₂	0.27	0.02	0.00	0.29
CZ	0.22	0.12	0.00	0.34
SCZ	0.05	0.30	0.80	1.15

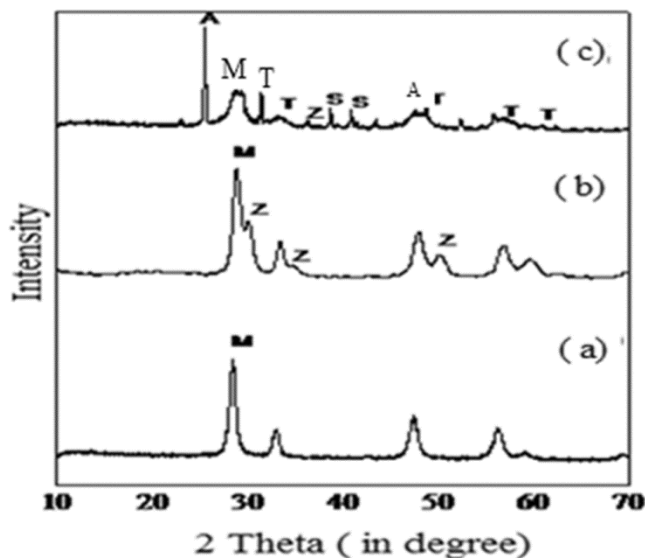


Fig. 2 — PXRD of HC coated with (a) Ceria, (b) CZ and (c) SCZ. [Ceria = M, Zirconia = Z, $\text{Zr}(\text{OH})_2\text{SO}_4 = \text{S}$, $\text{Zr}(\text{SO}_4)_2 = \text{A}$, T = tetragonal]

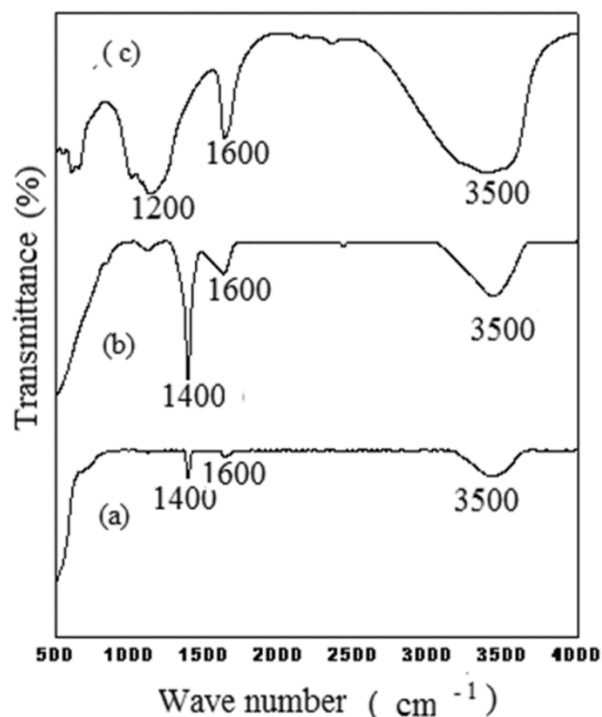


Fig. 3 — FT-IR spectrum: (a) Ceria, (b) CZ and (c) SCZ.

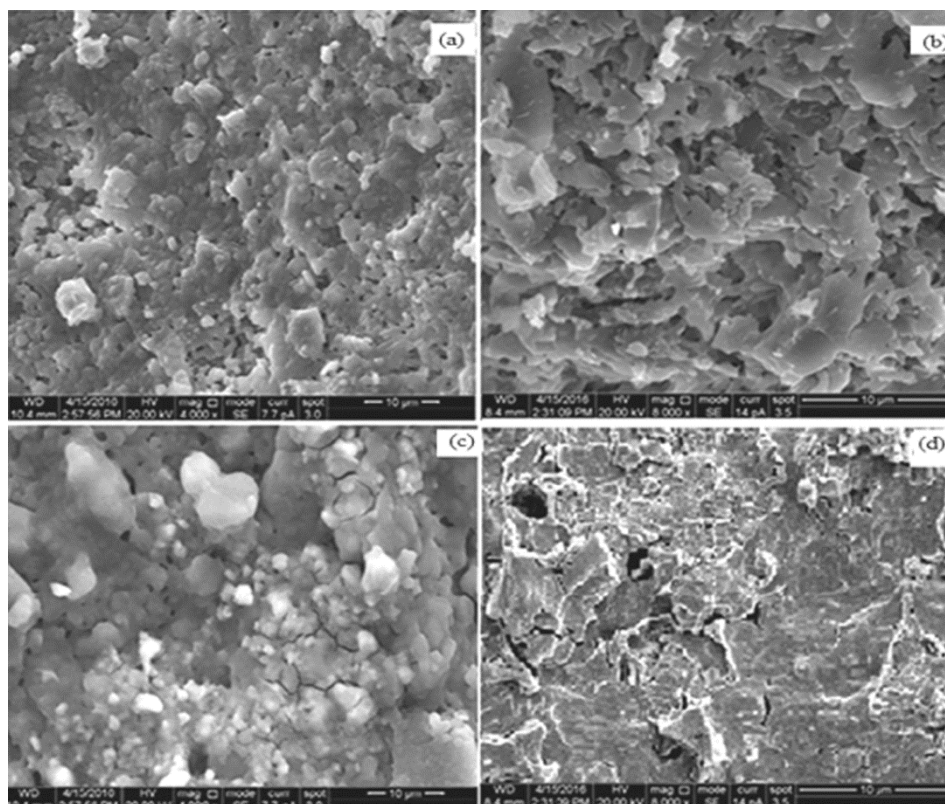


Fig. 4 — SEM image of honey comb coated solid acid catalysts (a) HC bare (b) Ceria (c) CZ, and (d) SCZ [Magnification = 4000 × 10 nm].

Reaction conditions-optimization studies

Optimization of reactions was carried out to get maximum yield of butyl acetate under microwave irradiation by varying the reaction conditions such as the nature of catalyst, molar ratio, reaction temperature and time.

Effect of nature of catalyst

Microwave-assisted esterification of butanol with acetic acid was conducted with different HC catalysts like ceria (C), ceria-zirconia (CZ) and sulfated-ceria – zirconia (SCZ). When there was no catalyst, the reaction occurs very slowly due to the weak acidity of acetic acid, consequently the formation of ester was negligible (8%). In the presence of the catalyst, the % yield of ester increases. The order of catalytic activity of various solid acid catalysts coated on HC is given below.

SCZ > CZ > C

The above order is similar to the acid sites determined from the NH₃-TPD method. The activity of HC catalyst in the synthesis of butyl acetate under microwave irradiation is enumerated in Table 2. Among all the catalyst, SCZ catalyst exhibits the highest catalytic activity and maximum yield of butyl acetate.

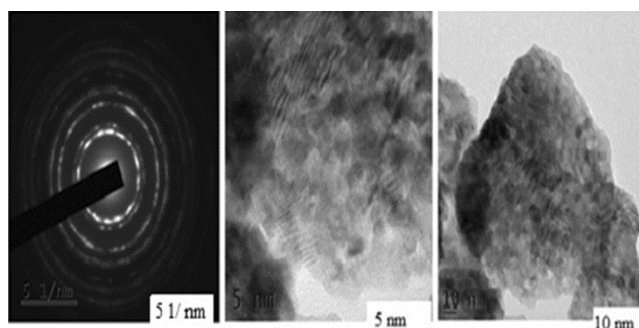


Fig. 5 — TEM images of SCZ at different magnification.

Table 2 — Synthesis of butyl acetate under microwave irradiation over different catalytic materials.

Catalyst	Yield of butyl acetate (%)	Conversion of butanol (%)
CeO ₂	50	60
CZ	78	85
SCZ	98	99

[Reaction conditions: Mole ratio of acetic acid: butanol = 1:2, Temperature= 80°C, reaction time= 12 min, catalyst weight= 0.15 g].

Effect of molar ratio

Molar ratios (acetic acid: *n*-butanol) were varied from 1:1 to 1:4 at 80°C by using SCZ- HC catalysts. The results are shown in Fig. 6.

Reduction of molar ratio decreases the ester yield, as a result of reducing the reactant concentration in the reaction mixture²⁵. Excess of alcohol favors the formation ester²⁶. When the concentration of one of the reactants like butanol increases from 1:1 to 1: 2 moles favours the formation of butyl acetate at a higher yield in accordance with Le Chatlier's Principle. When mole ratio increases beyond 1:2, % yield of butyl acetate reduces which may also result in blockage of active sites this may be due to the privileged adsorption of alcohol over the catalyst²⁷.

Effect of temperature

Effect of temperature on the esterification (acetic acid & *n*-butanol) was conducted at a different temperature, i.e., from 60°C to 100°C using SCZ catalyst coated on HC with a molar ratio of 1:2 for 12 min of 0.15 g catalyst. The obtained results are depicted as Fig. 7. From the graph, it was observed that, as the temperature increases formation of ester also increases due to the increase in the number of collisions per unit volume of acid to alcohol, maximum yield was obtained at 80°C. Formation of dibutyl ether as a side product at higher temperature is the reason behind the reduced % yield of butyl acetate^{27,28}.

Effect of reaction time

Synthesis of butyl acetate under microwave irradiation using SCZ catalyst was carried out at the different reaction time from 8 to 15 min. The results are enumerated in Fig. 8. From the graph it was observed that maximum yield of butyl acetate was obtained at 12 min. As the reaction time increases beyond 12 min (%) yield of butyl acetate decreases, due to the formation of the side product.

Techno-economical analysis

Techno-economical analysis of the catalysts is proposed by Liu, Weiping *et al*²⁹, used for the present work and briefly summarised in the following paragraph.

(a) Comparison of catalytic activity of catalyst by conventional vs microwave heating

In the present work synthesis of butyl acetate was carried out by both conventional and microwave heating overall honeycomb coated catalysts. It was noted that the (%) yield of butyl acetate obtained by conventional heating is low compared to microwave irradiation method and the results are given in Table 3.

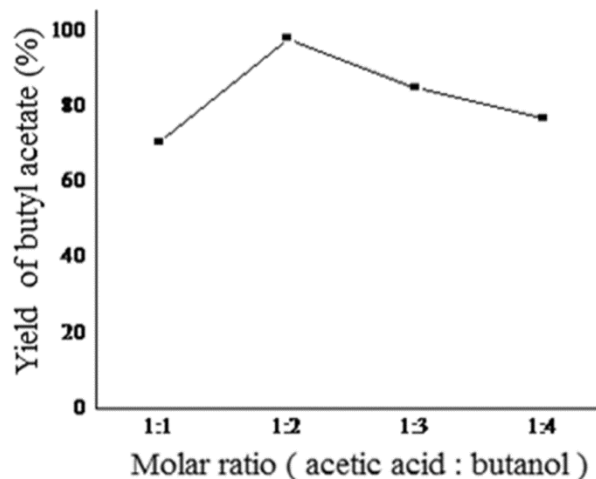


Fig. 6 — Effect of molar ratio of acetic acid: *n*-butanol. [Reaction condition: temperature = 80°C, reaction time =12 min, catalyst weight = 0.15 g].

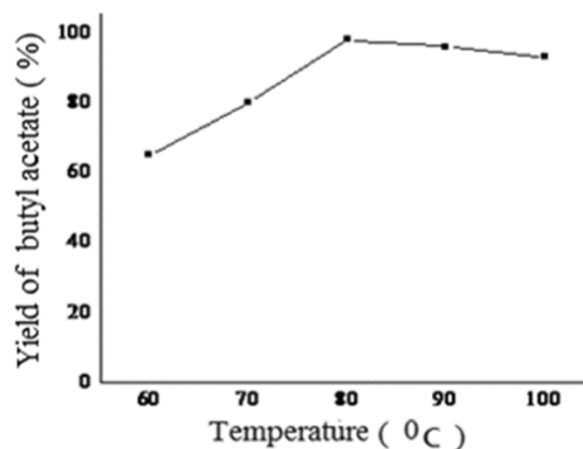


Fig. 7 — Effect of temperature [Reaction condition: Molar ratio of acetic acid :*n*-butanol= 1:2, reaction time =12 min, catalyst weight = 0.15 g].

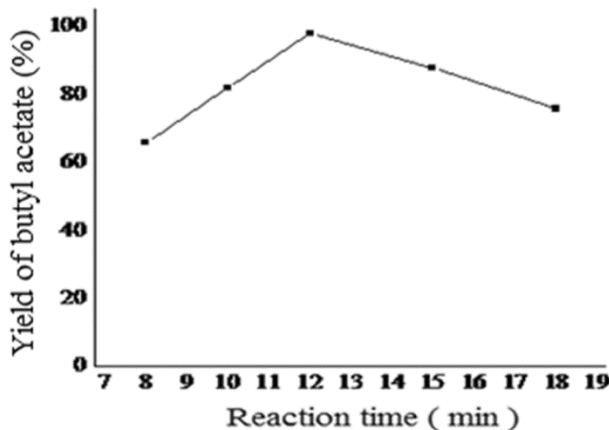


Fig. 8 — Effect of reaction time [Reaction condition: Molar ratio of acetic acid :*n*-butanol= 1:2, temperature = 80°C, catalyst weight = 0.15 g].

In presence of SCZ catalyst, 98% yield of butyl acetate was obtained under microwave irradiation in 12 min, whereas in conventional heating method only 20% of butylacetate was obtained. Further to obtain approximately 90% yield of butyl acetate under conventional heating it took nearly 6 h over SCZ catalyst. Hence microwave heating is more beneficial and economical than conventional heating.

(b) Comparison of catalytic activity of HC vs powder forms

Esterification reactions were conducted with butanol and acetic acid over catalysts in their powder and HC forms to asses of their catalytic activity. It was found that HC forms are more competent and economical than powder ones, although the same amount of the catalyst (~0.15 g) was used in the reaction (Table 4).

Honeycomb monolith has several advantages over powder catalyst such as excellent mass transfer, geometric surface area, trouble-free separation of products, low-pressure drop. In the case of HCs the thickness of the wall, the number of channels and their diameter evaluate the cell density. As cell density and surface area increases the catalytic performance of the catalyst also increases. HCs have a large open frontal area compared to powder form of the catalyst, the less amount of the catalyst loaded on HCs is compensated by the high efficiency because of the very good mass-transfer. Due to the homogenous dispersion of the catalyst, large number of active sites available on the surface of HC catalyst might increase the catalytic activity of the material, which is not possible in case of powder form³⁰.

The supremacy of the current work over other reported ones is evident by comparing our results with the results (Table 5). The reaction of butanol with acetic acid in presence sulfated-ceria-zirconia solid acid catalyst was chosen as a model reaction and the assessment was made in terms of ester yield and reaction temperature. Compared to other reported solid acids, sulfated-ceria-zirconia catalyst coated on honeycomb monolith leads to highest % yield of butyl acetate in a shorter reaction time and the catalyst can berecycled with no considerable loss in its catalytic activity even after five reaction cycles.

Compared to other solid acid catalysts, SCZ is having strong acidic sites. It is more stable, ecofriendly, easily separable, more efficient and economical too.

Reusability studies

The catalysts like CeO₂, CZ, and SCZ on both HC and powder form were studied for their reusability in

the microwave-assisted esterification reactions to synthesize butyl acetate (Table 6). After the first reaction cycle, the catalytic material in both the powder form and HC coated was removed from the reaction mixture, washed with acetone, dried in a hot air oven at 120°C for 2 h and calcined at 550°C for 1 h. It was noted that there is no change in the catalytic activity of all HCs in the esterification reaction even after successive five reaction cycles.

Table 3 — Comparison of catalytic activity of the catalysts (conventional v/s microwave heating).

Catalyst	Convention heating	Yield BA (%)	MW heating	Yield BA (%)
C	12 min	10	12 min	50
CZ	12 min	13	12 min	78
SCZ	12 min	20	12 min	98

[Reaction conditions: Molar ratio of acetic acid: butanol= 1:2, temperature = 80°C, catalyst weight = 0.15 g.]

Table 4 — Comparison of catalytic activity of HC v/s powder form of the catalysts.

Powder form	Yield BA (%)	HC Form	Yield BA (%)
CeO ₂	40 (3 h)	CeO ₂	50 (12 min)
CZ	70 (4 h)	CZ	78 (12 min)
SCZ	90 (6 h)	SCZ	98 (12 min)

[Reaction conditions: Molar ratio of acetic acid: butanol= 1:2, temperature= 80°C, reaction time= 12 min, catalyst weight= 0.15 g.]

Table 5 — Comparison of catalytic activity of SCZ catalyst with other reported solid acid catalysts.

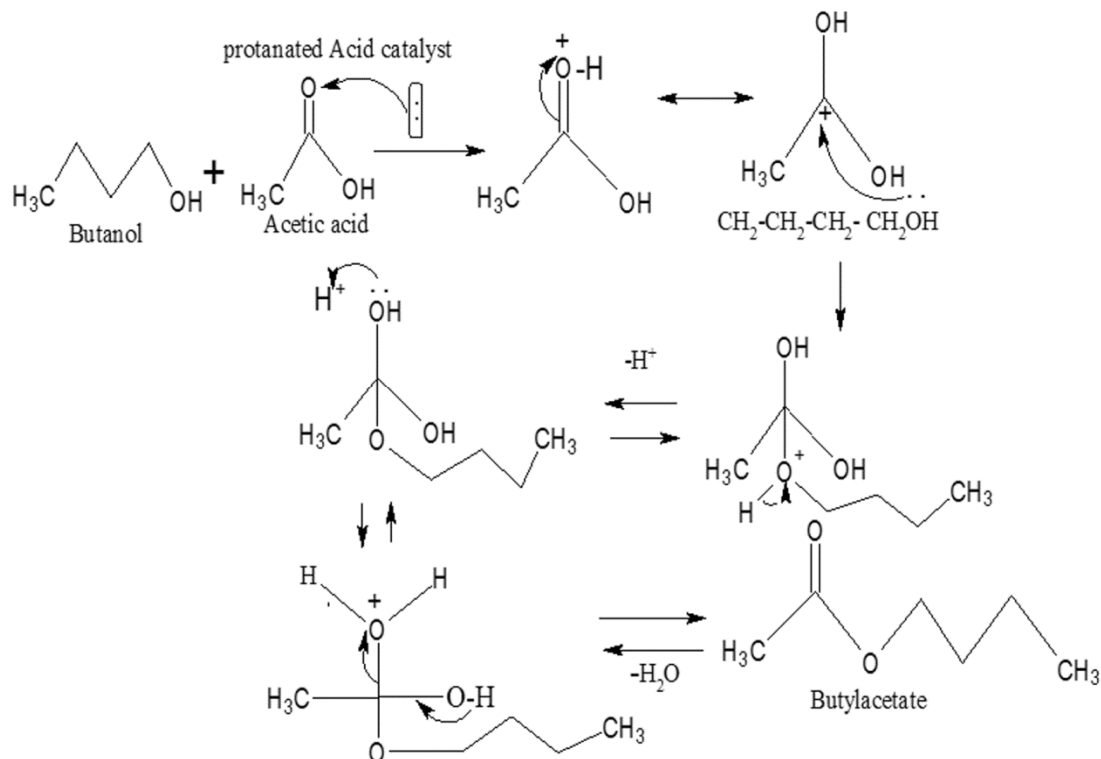
Sl. No.	Catalyst	Time	Yield (%)	Ref.
1	ZrO ₂ – WO ₃	-	85.6	31
2	Montmorillonite	15min	72.4	32
3	CZ	15min	64.1	33
4	MoO ₃ / SiO ₂ ²⁻	15min	91.0	34
5	Ion exchanged	30min	80.0	14
6	Montmorillonite SCZ	12 min	98.0	Present work

Table 6 — Reusability of solid acid catalysts.

Reaction cycles	Catalytic material					
	CeO ₂		CZ		SCZ	
	a	b	a	b	a	b
1	50	10	78	13	98	20
2	50	10	78	13	98	20
3	50	9	78	12	98	19
4	50	8	78	12	98	19
5	50	5	78	9	98	15

[Reaction conditions: Molar ratio of acetic acid: butanol= 1:2, temperature= 80°C, reaction time =12 min, catalyst weight= 0.15 g.]

Note: a - % of butyl acetate in HC coated catalyst; b - % of butyl acetate in powder form catalyst.



Scheme 1 — Mechanism for the synthesis of *n*-butyl acetate using acetic acid and *n*-butanol.

However, in the powder forms, the catalysts lose their catalytic activity when used for 5 reaction cycles. The more reusability of HCs can be because of the easy separation of the catalyst from the reaction mixture, which is impossible in powder forms.

Mechanism

The mechanism of the reaction takes places in the following steps.

Step-1 Protonated acid catalyst donates a proton to the carboxyl group of carboxylic acid to form carbocation.

Step-2 The nucleophile OH⁻ attacks the carbonation to form an intermediate compound, proton transfer, and loss of water followed by loss of the catalyzing proton to give ester³⁵.

Conclusion

The ceria based solid acid catalysts coated on honeycomb monoliths were found to be efficient as well as economical in the synthesis of butyl acetate. A correlation between the surface acidity and the catalytic activity of the catalysts was observed. The solid acid SO₄²⁻-CeO₂-ZrO₂ consisting of moderate and strong acid sites was found to be highly active in the synthesis of butyl acetate with 98% yield. The catalytic materials coated on honeycomb

monolith were found to be more efficient compared to the powder forms because they are easy to prepare, easy to use, remove, and are reusable. The % yield, molar ratio, and reaction timewere also investigated.

Acknowledgements

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