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# Catalytic performance of carbonaceous materials in the esterification of succinic acid

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## **Abstract**

Mesoporous carbonaceous materials have outstanding potential in many different applications such as adsorption, medicine and catalysis. We have recently reported the synthesis of a new form of mesoporous carbon, named Starbon<sup>®</sup>, obtained after low temperature carbonization of expanded starch. Such starch-derived mesoporous materials have several tunable properties including surface energies (ranging from hydrophilic to hydrophobic surfaces), which can be easily controlled by the degree of carbonization (from 200 to 700°C). Due to the diversity of surface functional groups, Starbons<sup>®</sup> can be easily chemically modified. Treatment of Starbon<sup>®</sup> materials with sulfuric acid gave a solid acid that has promising properties as heterogeneous catalyst. Comparative catalytic studies with some other similar commercial carbonaceous materials such as DARCO<sup>®</sup> and NORIT<sup>®</sup>, as well as phosphorous containing microporous carbons, are reported.

**Keywords:** Heterogeneous catalysis, carbonaceous materials, succinic acid esterification

## Introduction

Plants and animals are a source of many chemical products (metabolites), which can become key platform chemicals for synthesis, also known as “platform molecules”. Bio-platform molecules, also known as building blocks, are molecules with multiple functional groups that possess the potential to be transformed into new families of useful molecules, including commodity and specialty chemicals. Among them, the C<sub>3</sub> (glycerol and 3-hydroxypropionic acid), C<sub>4</sub> (Succinic (SA), Fumaric and Maleic acids, aspartic acid and 3-hydroxybutyrolactone), C<sub>5</sub> (levulinic, glutamic and itaconic acids) and C<sub>6</sub> (glucaric and 2,5-furan-di-carboxylic acid) have been ranked among the top platform chemicals, strengthened by a recent report by the US Department of Energy [1]. SA (1, 4-butanedioic acid) is a molecule of particular interest. It is a metabolite of the Krebs cycle, a respiratory process involving the breakdown of carbohydrates. Currently, SA is predominantly produced petrochemically. Maleic anhydride, produced via butane oxidation, is hydrated to maleic acid and then subsequently hydrogenated to SA. Production via this route exceeds 15,000 tpa. at a selling price between \$5.90-8.80/kg [2]. Nevertheless, we are now aware that SA, among several platform chemicals, can be produced from crops fermentation (after a few upstream processing stages), using different microorganisms in a bioreaction [3], hence fitting in the future paradigm of the Biorefinery. The versatile properties of SA [4] turn it into an important ingredient in the manufacture of various specialty and commodity chemicals including pharmaceuticals, plant growth stimulants, feed additives, green solvents, foods, detergents, surfactants, corrosion inhibitors and cosmetics as well as for the production of various commodity chemicals including esters, 1,4-butanediol (BDO), tetrahydrofuran, adipic acid,  $\gamma$ -butyrolactone, N-methylpyrrolidone, 2-pyrrolidone and succinate salts (Figure 1).

Of special interest, among all the products we can obtain from succinic acid, are the mono- and di-esters (esterification reactions) as they are important intermediates in the manufacture of polymers, fuel additives, solvents and personal care products [5]. The best way to achieve such intermediates and commodity chemicals should ideally be in a truly sustainable way, using chemical technologies with a low environmental impact. This includes the use of green chemistry methods such as heterogeneous catalysis [6, 7] and the application of green chemistry principles [8].

In a greener approach to all different acid-catalysed chemical processes, the use of recyclable solid acid materials as replacement for the typically homogeneous acid catalysed conditions has been recently promoted [7, 9, 10]. We have recently reported that carbonaceous materials were found to be the best candidates, among solid acids, in various acid catalysed reactions [11, 12, 13]. Few authors have reported carbonaceous carbohydrate systems in catalytic applications [14, 15] and the reported active materials were mostly microporous in nature.

The develop of the novel mesoporous Starbon® materials opened up exciting opportunities for the conversion of larger molecules into valuable petrochemicals and fine chemicals. A whole range of polysaccharide derived mesoporous materials from starch to activated carbon, including amorphous oxygen containing carbons, can be easily synthesized controlling the degree of carbonization, and have many applications owing to their varied surface functionalities.

There is a need to study and compare activities of the different carbonaceous materials (microporous versus mesoporous) in various reactions, in order to have a better knowledge and understanding of the influence of the materials properties in the catalytic performance.

Here we report the comparison of activities for several acidic carbonaceous materials in the esterification of succinic acid in water.

## **Experimental**

### *Materials synthesis*

The Starbon® synthesis procedure has been previously reported [11]. Figure 2 shows the typical mass loss profiles during Starbon® preparation together with differential thermogravimetric (DTG) curves of different temperature-prepared Starbons®. Of note is that native starch granules do not produce mesoporous materials when carbonized. This indicates that the formation of expanded starch as a precursor to Starbon® is crucial. As synthesized materials were suspended in H<sub>2</sub>SO<sub>4</sub> 99.999% purity (10 mL acid g<sup>-1</sup> material) and heated for 4 h at 80°C. After sulphonation, samples were subsequently washed with distilled water (to flush out the residual non adsorbed acid)

until the washings were neutral, and oven dried overnight at 100°C. Starbon® acids were then extracted with toluene (4 h in boiling toluene), and water (3 h, 80°C) and finally oven dried (100°C) overnight before being tested in the catalytic reaction.

The preparation of the Starbon® materials is reproducible (Table 1). As-synthesized materials prepared in different batches showed similar textural properties to those of the Starbon®-400-SO<sub>3</sub>H included in Table 2. The sulphonation of the materials did not significantly change such textural properties.

Sulphonated commercial microporous DARCO® and NORIT® were synthesized from the microporous commercial carbons in a similar way.

Phosphorous-containing carbons [referred to as Carbon-P and (A)Carbon-P (activated)], respectively, were prepared from porous spherical phenol-formaldehyde resin [16] phosphorylated by refluxing in a phosphorous oxychloride solution in toluene. The derived precursor was then carbonised in three steps at 250 [Carbon-P-250 and (A)Carbon-P-250], 500 [Carbon-P-500 and (A)Carbon-P-500] and 750°C [Carbon-P-750 and (A)Carbon-P-750] in a flow of oxygen-free nitrogen. Materials were then washed with water, a solution of 0.1 M NaOH, water, a solution of 0.1 M HCl and water and finally oven dried (120°C) until constant weight was achieved (Carbon-P samples). For the activated samples, (A)Carbon-P, materials were prepared after oxidation with hydrogen peroxide.

### *Characterisation*

Thermogravimetric analysis (TG) was performed using a Netzsch STA 409 at scan rates of 10°C min<sup>-1</sup>, with typically 20mg sample under flowing N<sub>2</sub> at 100 mL min<sup>-1</sup>.

Thermogravimetric analysis coupled with infrared (TG-IR) was performed using a Netzsch STA 409 at scan rates of 10°C min<sup>-1</sup>, with typically 20mg sample under flowing N<sub>2</sub> at 100 mL min<sup>-1</sup>, coupled with a Brüker EQUINOX-55 instrument equipped with a liquid N<sub>2</sub> cooled MCT detector. This technique was employed to measure the materials sulfur loading.

Nitrogen adsorption measurements were carried out at 77 K using an ASAP2010 volumetric adsorption analyser from Micromeritics. Samples were outgassed prior

measurement under vacuum for 3-4 h at the temperature equal to temperature of sample preparation. Materials surface acidity was measured in a dynamic mode by means of the gas phase adsorption of pyridine (PY) using a pulse chromatographic technique [17] and by pH-titration with 0.01 M NaOH and 0.5 M HCl in the presence of 0.1 M NaCl.

#### *Catalytic test*

A typical catalytic test was performed as follows: 1 mmol Succinic acid (SA, 0.124 mg), 30 mmol EtOH (2.4 mL) and 50 mmol water (0.9 mL) were added to a round bottom flask with 0.1 g of solid acid catalyst, increasing the temperature to 80°C. Samples were withdrawn periodically from the reaction mixture and the mixture was left reacting for 24 hours. Products were analysed by GC using an Agilent 6890N GC model equipped with a 7683B series autosampler. Response factors of the reaction products were determined with respect to succinic acid from GC analysis using known compounds in calibration mixtures of specified compositions. Three main products were found from the reaction mixture: succinic anhydride (SANH), monoethyl succinate (MES) and diethyl succinate (DIES), respectively.

## **Results and Discussion**

#### *Carbonaceous materials properties*

Table 2 summarises the textural [surface area ( $S_{\text{BET}}$ ), pore diameter ( $D_{\text{BJH}}$ ), pore volume ( $V_{\text{BJH}}$ )] and acid properties ( $\text{HSO}_3$ -loading, surface acidity) of the different micro-mesoporous carbonaceous materials synthesized.

The phosphorous content in the Carbon-P samples was approximately 7% measured either by elemental analysis or by the molybdate blue method [18]. The (sulphonated) Starbon® was found to have the typical textural porosity at the upper limit of the mesoporous domain with an increase in the microporous region for materials heated at temperatures higher than 500°C, as previously reported [11]. A comparison between the different carbonaceous materials (prior to sulphonation) is shown in Figure 3. The microporous volume was found to decrease in the materials from DARCO® (A) to

Starbon® (D) with the mesoporous region remaining almost unchanged. The sulphonated carbons exhibited similar isotherm profiles to those of the parent materials.

In terms of surface acidity, the commercial sulphonated DARCO® and NORIT®, together with the sulphonated Starbon® were highly acidic compared with the non activated carbon-P materials synthesized (Table 1). The ion exchanged properties of the microporous carbon-P materials, determined by pH-titration, showed a flat curve with no significantly defined inflections, indicating a weakly acid functionality [18]. Nevertheless, the negative surface charge found in the materials, attributed to the dissociation of acidic phosphorous-containing groups on the materials surface, are likely to increase the surface acidic properties, rendering materials that are more acidic than some oxidized active carbons [18, 19]. Activated carbon-P materials exhibited similar acidity to that of the Starbon® acid and the sulphonated DARCO® and NORIT®.

#### *Catalytic activity*

We recently reported the catalytic activity of sulphonated Starbons® in the esterification of succinic acid in aqueous ethanol (Figure 4). [12] Materials were shown to be very active and selective in the production of the mono/di-ester. A range of carbonaceous materials were then tested in the esterification reaction and compared to the Starbon® acids. Results are summarized in Table 3.

The esterification of succinic acid is an autocatalytic reaction and consequently the reaction progresses by itself without the need of any catalyst. Nevertheless, the autocatalytic reaction is extremely slow, taking 48 hours to complete.

The low to high temperature non activated carbon-P materials exhibited similar catalytic activities in the esterification reaction, consistent with the surface acidity data (Table 2). The most acidic catalysts [(A)Carbon500 and (A)Carbon750, respectively] showed the best results in terms of succinic acid conversion and selectivity to the DIES, in good agreement, again, with the titration data.

Sulphonated DARCO® and NORIT® displayed a similar performance in the esterification compared to the phosphorous-containing carbons (Table 3), although an improved selectivity to the DIES was found, probably due to the higher acidity (in particular with respect to non-activated carbon-P materials) together with pore

distribution/surface properties. However, all of the reactions performed using the carbonaceous solid acids were very slow and quantitative conversion of the substrate took 12+ hours at 80°C, with the notable exception of the Starbon®-400-HSO<sub>3</sub>. The reported catalytic activities of the Starbon® acids are reproducible from batch to batch.

Despite having similar acidities compared to the activated carbon-P and the microporous carbonaceous materials, the rates of reaction for the Starbon® acid catalyst were found to be at least 5 times greater than any of the other solid acid catalysts screened in the esterification, enabling the quantitative conversion of the acid within 3-4 h and quantitative production of the diester within 5 h (Table 3). Moreover, the catalyst was easily recovered from the aqueous reaction mixture whereupon the reaction rates return to the background values. The recovered Starbon® could be added to fresh substrate solutions giving almost identical reaction rates to that observed in the original reaction. Thus the catalyst is stable as well as highly active under aqueous conditions, most unusual characteristics. Indeed, water has been reported to decrease the esterification reaction rate, a phenomenon that has been attributed not only to a reverse hydrolysis [20, 21] but also to a competitive protonation step involving the water and the alcohol (assuming the rate-determining step is the interaction between the protonated alcohol and the carboxylic acid) [22]. The effect of water on the esterification of acids under homogeneous conditions has been recently reported. Sulfuric acid provides a very fast rate of conversion of the acid, but its catalytic activity is strongly inhibited by water (losing up to 90% activity with increasing water concentrations) [23]. Our studies prove that while sulfuric acid provided a very fast rate of conversion of the acid, the rate of formation of the diester is considerably slower than with any of the sulphonated materials (Table 3).

We believe that different hydrophilic/hydrophobic environments within the most active Starbon® acids catalyst pores provide an ideal combination of properties in terms of hydrophobicity, encouraging partitioning of the organic acid into the pores and production of the more hydrophobic esters. The material mesoporosity also enhances molecular diffusion rates.

## Conclusions

By applying green chemical technologies to the transformation of widely available platform molecules, we can build up a new sustainable chemical industry for the 21<sup>st</sup> century. In this regard, we have demonstrated that carbonaceous materials are the best candidates to carry out esterifications of diacids in aqueous solution. A comparative study of various micro-mesoporous carbonaceous materials showed the mesoporous Starbon® acids as 2-10 times more active than microporous commercially available carbonaceous materials and synthetic phosphorous containing carbons in the esterification reaction, despite the similar acidities and mesoporous volume. The high activity of the Starbon® acids is believed to be due to a combination of the materials specific hydrophilic/hydrophobic environments and the microporosity/mesoporosity ratio. The catalysts were also reusable.

## Acknowledgements

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Table 1. Textural properties of Starbon® materials prepared in different batches.

Batch	$S_{\text{BET}}$ ( $\text{m}^2 \text{g}^{-1}$ )	$D_{\text{BJH}}^{\text{ads}}$ (nm)	$V_{\text{BJH}}$ ( $\text{cm}^3 \text{g}^{-1}$ )
1	386	4.9	0.53
2	332	4.8	0.52
3	393	4.6	0.53
4	411	3.9	0.58

Table 2. Textural (surface area,  $S_{\text{BET}}$ , pore diameter,  $D_{\text{BJH}}$ , and pore volume,  $V_{\text{BJH}}$ ) and surface acid properties ( $\text{HSO}_3$  loading,  $\text{mmol g}^{-1}$ , and surface acidity,  $\text{mmol g}^{-1}$  of Py at  $200^\circ\text{C}$ / NaOH neutralization capacity,  $\text{mmol g}^{-1}$ ) of the different carbonaceous materials.<sup>a</sup>

Catalysts	$S_{\text{BET}}$ ( $\text{m}^2 \text{g}^{-1}$ )	$D_{\text{BJH}}$ (nm)	$V_{\text{BJH}}$ ( $\text{cm}^3 \text{g}^{-1}$ )	$\text{HSO}_3$ loading ( $\text{mmol g}^{-1}$ )	Surface acidity ( $\text{mmol g}^{-1}$ )
Starbon®-400- $\text{HSO}_3$	386	4.1	0.62	0.5	205 <sup>a</sup>
DARCO®- $\text{HSO}_3$	585	0.6	1.47	0.3	190 <sup>a</sup>
NORIT®- $\text{HSO}_3$	626	0.5	1.24	0.3	198 <sup>a</sup>
Carbon-P-250	365	0.6	0.21	-	93 <sup>a</sup> /0.88 <sup>b</sup>
(A)Carbon-P-250	279	0.7	0.19	-	178 <sup>a</sup> /2.32 <sup>b</sup>
Carbon-P-500	430	0.7	0.20	-	124/0.96 <sup>b</sup>
(A)Carbon-P-500	372	0.7	0.18	-	200/2.14 <sup>b</sup>
Carbon-P-750	450	0.7	0.13	-	108 <sup>a</sup> /0.84 <sup>b</sup>
(A)Carbon-P-750	396	0.6	0.18	-	192 <sup>a</sup> /1.72 <sup>b</sup>

<sup>a</sup>Pyridine (PY) titration value at  $200^\circ\text{C}$ ; <sup>b</sup>measured as NaOH neutralization capacity ( $\text{mmol g}^{-1}$ )

Table 3. Time of reaction (h), total conversion ( $X_T$ , mol% SA converted) and selectivities to the monoester ( $S_{MES}$ , mol%), diester ( $S_{DIES}$ , mol%) and anhydride ( $S_{SANH}$ , mol%) for the esterification of succinic acid with ethanol over porous carbonaceous materials.<sup>a</sup>

Catalyst	Time (h)	$X_T$ (mol% SA)	$S_{MES}$ (mol%)	$S_{DIES}$ (mol%)	$S_{SANH}$ (mol%)
No catalyst	4	24	80	10	10
	24	60	65	35	-
$H_2SO_4$ cc	4	>95	33	67	-
	24	>95	10	90	-
Starbon®-400- $HSO_3$	1	52	71	22	7
	4	90	20	80	-
	8	>95	<5	>95	-
DARCO®- $HSO_3$	4	60	78	18	4
	8	80	68	27	5
	20	>95	10	90	-
NORIT®- $HSO_3$	8	70	68	29	3
	12	>95	45	55	-
Carbon-P-250	4	52	79	21	-
	24	>95	60	40	-
(A)Carbon-P-250	4	74	68	23	9
	12	>95	62	38	-
Carbon-P-500	4	60	78	16	6
	24	>95	51	49	-
(A)Carbon-P-500	4	78	70	23	7
	12	>95	41	59	-
Carbon-P-750	4	42	76	14	10
	24	>95	71	25	4
(A)Carbon-P-750	4	85	69	16	15
	12	>95	50	47	3

<sup>a</sup>Reaction conditions: 1 mmol Succinic acid (SA, 0.124 mg), 30 mmol EtOH (2.4 mL) and 50 mmol water (0.9 mL), 80°C.

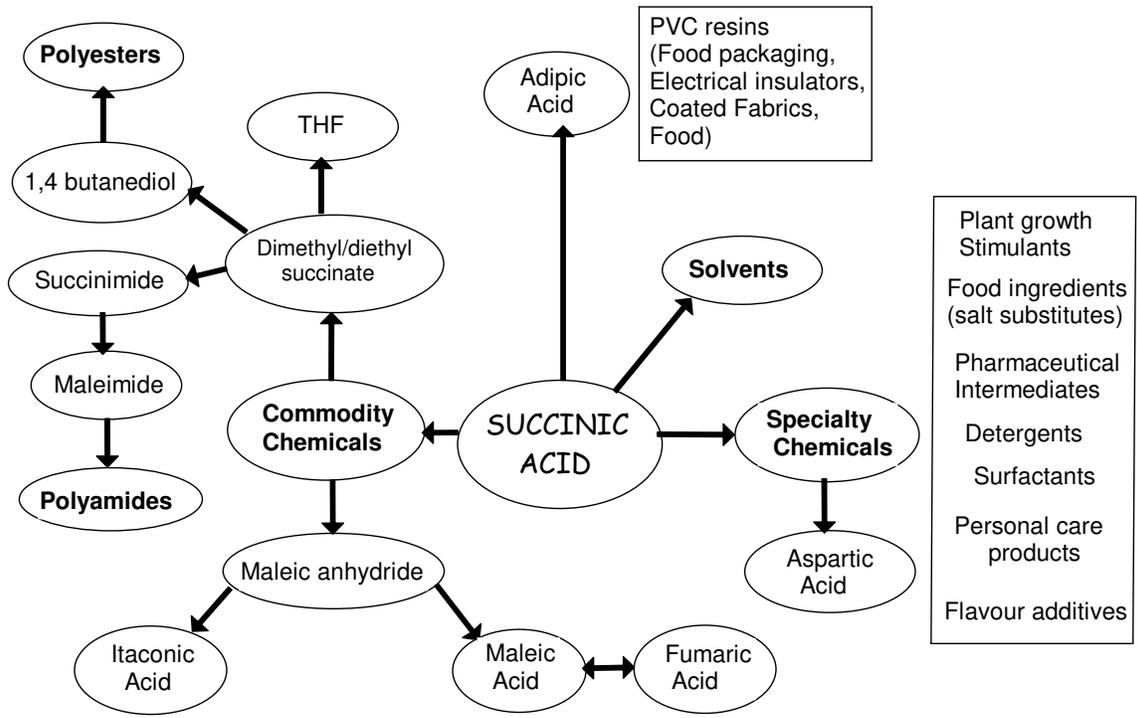


Figure 1. Master plan for the development of Succinic Acid-based products: from specialty to commodity chemicals.

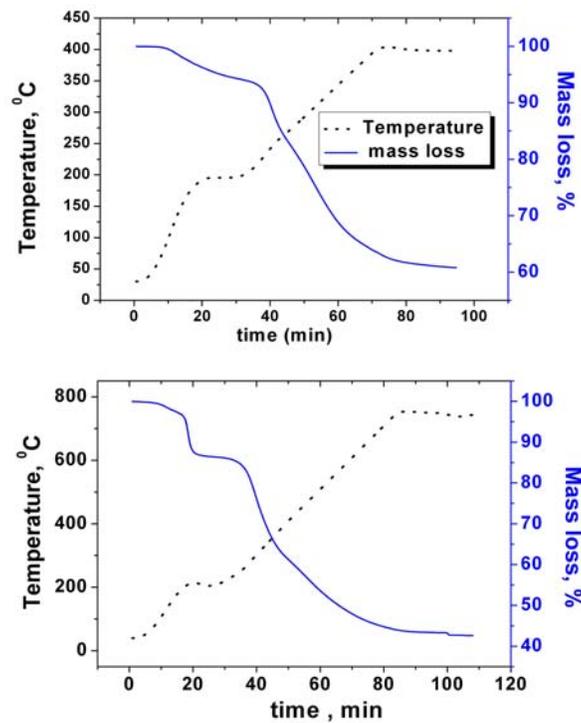


Figure 2. Typical thermogravimetric mass loss profiles (wt%) during Starbon®400 (top) and Starbon®750 (bottom) preparation (Temperature vs time).

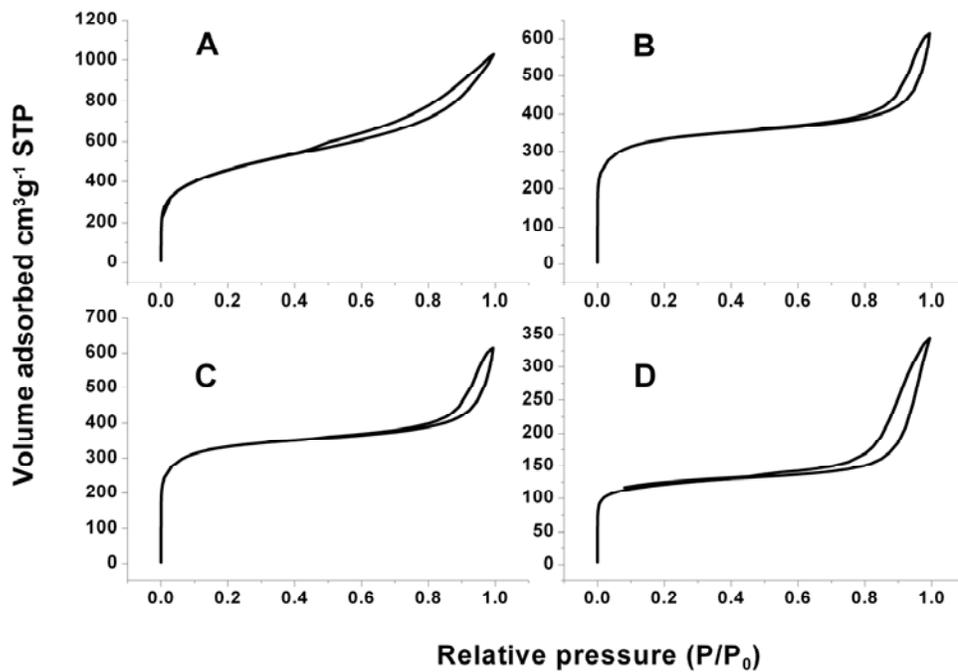


Figure 3. Nitrogen adsorption-desorption isotherm profiles of various carbonaceous materials. A) DARCO®; B) NORIT®; C) (A)Carbon-P-500; D) Starbon®-400.

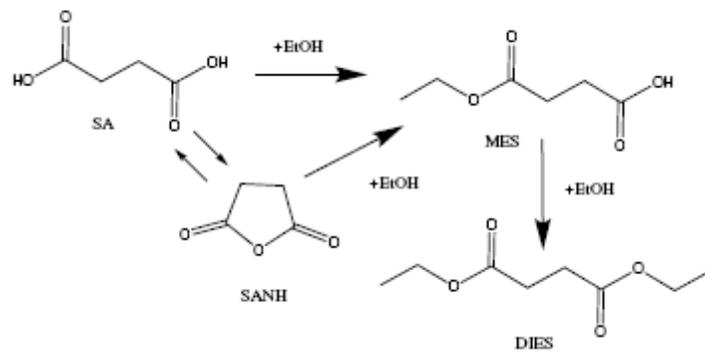


Figure 4. Products obtained in the esterification reaction of succinic acid