There remains a need for cost-effective chemical recycling of PLA and successful integration of the hydrolyzed PLA with existing LA/PLA production processes.

There also remains a need to improve the yield of lactic acid production, particularly from organic waste.

SUMMARY

In an aspect, the present disclosure refers to a method for producing an L-lactate salt from a combined recycling of polylactic acid (PLA) waste and recycling of organic waste, the method comprising the steps of:

(a) hydrolyzing PLA waste comprising poly-L-lactic acid (PLLA), poly-D-lactic acid (PDLA), poly (D-L-)-lactic acid (PDLLA), stereocomplex PLA (sc-PLA) or a combination thereof with a metal oxide or a hydroxide to obtain a PLA hydrolysis slurry comprising L- lactate monomers and a first counterion;

(b) fermenting organic waste comprising carbohydrates and selected from a group consisting of food waste, organic fraction of municipal waste, agricultural waste, plant material, and a combination thereof with a lactic acid-producing microorganism in a fermenter in the presence of an alkaline compound to obtain a fermentation broth comprising L- lactate monomers and a second counterion,

wherein the first and second counterions are the same; or wherein at least one of the first and second counterions is subjected to ion exchange thereby obtaining a first and second counterions which are the same;

(c) optionally contacting the PLA hydrolysis slurry of step (a), the fermentation broth of step (b), or a mixture comprising the PLA hydrolysis slurry of step (a) and the fermentation broth of step (b) with a D-lactic acid degrading enzyme or a D-lactic acid-utilizing microorganism to eliminate D-lactate monomers thereby obtaining L-lactate monomers; and

(d) purifying a mixture comprising the PLA hydrolysis slurry of step (a) and the fermentation broth of step (b) or the L-lactate monomers of step (c) thereby obtaining an L-lactate salt.

The present disclosure provides methods and systems for highly efficient production of an enantiomerically pure lactate salt, particularly an L-lactate salt, by combined recycling of organic waste and PLA waste. More particularly, the present disclosure integrates production of L-lactate monomers by lactic acid fermentation of organic waste with chemical hydrolysis of PLA to its constituting monomers (L- and optionally D-lactate monomers). As disclosed herein, L-lactate monomers produced by fermentation and lactate monomers produced by chemical hydrolysis of PLA are combined and purified together in a single downstream purification and recovery process to obtain a pure L-lactate salt. In some examples, the lactate monomers resulting from PLA hydrolysis are combined with L-lactate monomers produced by fermentation after the fermentation is completed. Alternatively, lactate monomers produced by PLA hydrolysis are supplemented to a lactic acid production reactor in which L-lactate monomers are produced by fermentation of organic waste, and subsequently the supplemented lactate monomers and newly produced L-lactate monomers are subjected to a single purification process to obtain a pure L-lactate salt. The purified L-lactate salt can then be acidified into L-lactic acid and used in the production of new PLA.

As disclosed herein, lactic acid fermentation is carried out in the presence of an alkaline compound that adjusts the pH during fermentation, resulting in a fermentation broth comprising L-lactate monomers and a counterion. The PLA hydrolysis is carried out using a metal oxide or a hydroxide, resulting in a hydrolysis slurry comprising lactate monomers (L- and optionally D-) and a counterion. The alkaline compound used during fermentation and the metal oxide or the hydroxide used for PLA hydrolysis according to the present disclosure produce L-lactate monomers and a counterion which may be the same or different with each possibility representing a separate example. When using the same counterion, the L-lactate monomers and the counterions can be combined and purified together to obtain a pure lactate salt. When using a different counterion, at least

one of the counterions can be exchanged thereby resulting in the same counterion which together with the L-lactate monomers can be subjected to subsequent combined purification. In some examples, the alkaline compound used during fermentation and the metal oxide or the hydroxide used for PLA hydrolysis are the same compound, namely, the same compound is used for both PLA hydrolysis and pH adjustment in the fermenter. For example, magnesium hydroxide can be used both as a hydroxide for PLA hydrolysis and as an alkaline compound for pH adjustment during fermentation, resulting in lactate monomers and magnesium ions in both the hydrolysis slurry and the fermentation broth, which can be recovered as magnesium lactate. In other examples, the compounds are different, but produce the same counterion. For example, magnesium hydroxide can be used as a hydroxide for PLA hydrolysis, and magnesium carbonate can be used as an alkaline compound for pH adjustment during fermentation, resulting in lactate monomers and magnesium ions in both the hydrolysis slurry and the fermentation broth, which can be recovered as magnesium lactate. In yet other examples, the compounds are different, but one of the counterions is exchanged to produce the same counterion as the other for subsequent purification. For example, sodium hydroxide can be used as a hydroxide for PLA hydrolysis, and magnesium hydroxide can be used as an alkaline compound for pH adjustment during fermentation. The sodium ions in the hydrolysis slurry can then be exchanged with magnesium ions thereby resulting in lactate monomers and magnesium ions in both the hydrolysis slurry and the fermentation broth, which can be recovered as magnesium lactate salt. In further examples, both counterions are exchanged to produce the same counterion for subsequent purification.

The present invention advantageously integrates the products of two processes, namely, i) lactate hydrolyzed from PLA and ii) lactate produced via fermentation of organic waste, into a single downstream purification process for recovering an L-lactate salt, thus saving both capital expenditure (CAPEX) and operational expenditure (OPEX).

In addition, the present invention improves the yield of L-lactic acid production from organic waste. Organic waste such as mixed food waste is characterized by high variability in terms of carbohydrate content, and its composition varies from batch to batch where some batches may be rich in carbohydrates compared to others that have a low carbohydrate content. Thus, lactic acid fermentation processes based on organic waste may result in varied yields from batch to batch, and in certain instances a very low

yield of lactic acid is obtained. The integration of lactate hydrolyzed from PLA and lactate produced via fermentation of organic waste increases the amount of lactic acid that is obtained per fermentation cycle, thus improving the yield and facilitating reproducibility of lactic acid fermentation processes.

The systems and methods of the present disclosure therefore provide recycling of PLA and organic waste which is cost-effective and improves the yield of L-lactic acid production.

According to a first example, there is provided a method for producing an L-lactate salt from a combined recycling of polylactic acid (PLA) and organic waste, the method comprising the steps of:

(a) hydrolyzing PLA waste with a metal oxide or a hydroxide to obtain a PLA hydrolysis slurry comprising L- lactate monomers and a first counterion;

(b) fermenting organic waste with a lactic acid-producing microorganism in a fermenter in the presence of an alkaline compound to obtain a fermentation broth comprising L- lactate monomers and a second counterion,
wherein the first and second counterions are the same; or wherein at least one of the first and second counterions is subjected to ion exchange thereby obtaining a first and second counterions which are the same;

(c) optionally contacting the PLA hydrolysis slurry of step (a), the fermentation broth of step (b), or a mixture comprising the PLA hydrolysis slurry of step (a) and the fermentation broth of step (b) with a D-lactic acid degrading enzyme or a D-lactic acid-utilizing microorganism to eliminate D-lactate monomers thereby obtaining L-lactate monomers; and

(d) purifying a mixture comprising the PLA hydrolysis slurry of step (a) and the fermentation broth of step (b) or the L-lactate monomers of step (c) thereby obtaining an L-lactate salt.

In one example, step (a) and step (b) are performed in any order or simultaneously, with each possibility representing a separate example.

In some examples, the PLA hydrolysis slurry is gradually added to the lactic acid fermenter of step (b) during the fermentation. In accordance with these examples, the mixture comprising the PLA hydrolysis slurry of step (a) and the fermentation broth of step (b) is obtained by gradually adding the PLA hydrolysis slurry to the lactic acid

fermenter during fermentation. In some examples, where the PLA hydrolysis slurry is added to the lactic acid fermenter of step (b), step (a) may comprise the metal oxide or the hydroxide in excess thereby obviating the need to add the alkaline compound to the lactic acid fermenter in step (b) for pH adjustment during fermentation.

Advantageously, the integration of the L-lactate monomers produced by lactic acid fermentation with the L-lactate monomers resulting from PLA hydrolysis into a single downstream purification process to obtain a purified L-lactate salt increases the overall yield of L-lactate production, which is particularly beneficial for organic wastes with a low carbohydrate content that cannot reach high titers of lactic acid. In one example, the overall yield of L-lactate production is increased by 10% or more. In another example, the overall yield of L-lactate production is increased by 50% or more. In yet another example, the overall yield of L-lactate production is increased by 100% or more.

In certain examples, the lactate salt is magnesium L-lactate. In particular examples, the lactate salt is crystalline magnesium L-lactate. In specific examples, the lactate salt is crystalline magnesium L-lactate dihydrate.

In some examples, the metal oxide or hydroxide of step (a) and the alkaline compound of step (b) are the same compound.

In various examples, the alkaline compound of step (b) is a metal oxide, a carbonate or a hydroxide. Each possibility represents a separate example.

In certain examples, the metal oxide comprises at least one of MgO, CaO and a mixture or combination thereof. Each possibility represents a separate example.

In other examples, the carbonate comprises at least one of $CaCO_3$, $MgCO_3$ and a mixture or combination thereof. Each possibility represents a separate example.

In certain examples, the hydroxide comprises at least one of NaOH, KOH, NH_4OH, $Ca(OH)_2$, $Mg(OH)_2$ and a mixture or combination thereof. Each possibility represents a separate example.

In one particular example, the hydroxide of step (a) is NaOH and the alkaline compound of step (b) is $Mg(OH)_2$. In another particular example, the hydroxide of step (a) and the alkaline compound of step (b) are $Mg(OH)_2$.

In various examples, the hydrolysis in step (a) is performed at elevated temperatures in the range of about 50°C to about 90°C, for example, temperatures in the

range of about 60°C to about 90°C, about 70°C to about 90°C, about 50°C to about 80°C, and about 50°C to about 75°C, including each value within the specified ranges. Each possibility represents a separate example.

In further examples, the hydrolysis in step (a) is performed for a period of time in the range of about 1 to about 12 hours, including each value within the specified range. In other examples, the hydrolysis in step (a) is performed for a period of time in the range of about 12 to about 36 hours, including each value within the specified range.

In additional examples, step (a) comprises hydrolyzing PLA waste with $Mg(OH)_2$ at a concentration of between about 5 to about 15 wt.% to obtain a PLA hydrolysis slurry comprising L- lactate monomers and a magnesium ion.

In other examples, the hydrolysis in step (a) results in L- lactate monomers and a first counterion in crystalline form. In yet other examples, the fermentation in step (b) results in L-lactate monomers and a second counterion in crystalline form.

In additional examples, the method for producing an L-lactate salt further comprises pretreatment of the PLA waste prior to step (a). In specific examples, pretreatment comprises a mechanical pretreatment selected from the group consisting of grinding, chipping, shredding, milling, and a combination thereof. Each possibility represents a separate example. In other specific examples, pretreatment comprises extrusion pretreatment.

In further examples, the method for producing an L-lactate salt further comprises subjecting the PLA hydrolysis slurry obtained in step (a) to solid-liquid separation. It is contemplated that the solid-liquid separation provides the removal of unhydrolyzed PLA waste or impurities such as other polymers, inert materials and/or food waste from the slurry.

PLA waste according to the present invention may include non-PLA impurities and contaminants. In some examples, the PLA waste is sorted prior to step (a) to increase the amount of PLA in said waste relative to non-PLA impurities and contaminants. Advantageously, PLA recycling according to the present invention is insensitive to impurities and contaminants present in the PLA waste, including contaminants that cannot be sorted successfully. As disclosed herein, the PLA waste following the alkaline hydrolysis is integrated into a downstream purification process of a lactic acid fermentation broth, the downstream purification process simultaneously eliminates

contaminants originating from both the organic waste that was used as the substrate for fermentation and the PLA waste.

In certain examples, the PLA waste comprises both poly L-lactic acid (PLLA) and poly D-lactic acid (PDLA).

In some examples, the organic waste comprises endogenous D-lactic acid, L-lactic acid or a combination thereof. Each possibility represents a separate example.

In certain examples, where D-lactic acid is formed and/or present, the PLA hydrolysis slurry of step (a), the fermentation broth of step (b) or a mixture comprising the PLA hydrolysis slurry of step (a) and the fermentation broth of step (b) are contacted with a D-lactic acid degrading enzyme or a D-lactic acid-utilizing microorganism to eliminate D-lactate monomers thereby obtaining L-lactate monomers. In particular examples, the D-lactic acid degrading enzyme in step (c) is a D-lactate oxidase.

In some examples, the organic waste is selected from the group consisting of food waste, municipal waste, agricultural waste, plant material and a mixture or combination thereof. Each possibility represents a separate example.

In other examples, the obtained L-lactate salt is purified by at least one of crystallization, recrystallization, distillation, partitioning, silica gel chromatography, preparative HPLC, and combinations thereof. Each possibility represents a separate example.

In further examples, the obtained L-lactate salt is acidified to form L-lactic acid by at least one of hydrochloric acid, hydrobromic acid, phosphoric acid, sulfuric acid, and combinations thereof. Each possibility represents a separate example. In particular examples, the L-lactic acid is used for subsequent polylactic acid formation.

According to a second example, there is provided a method for producing magnesium L-lactate salt by recycling of polylactic acid (PLA) waste, the method comprising the steps of:

(a) hydrolyzing PLA waste with a base selected from sodium, potassium and ammonium hydroxide to obtain a PLA hydrolysis slurry comprising L- lactate monomers and a counterion selected from sodium, potassium and ammonium;

(b) optionally performing at least one of neutralizing the PLA hydrolysis slurry with an acid and removing unhydrolyzed PLA waste; and

(c) adding a magnesium salt to the PLA hydrolysis slurry of step (a) or (b) to thereby precipitate magnesium L-lactate salt.

In some examples, the base is sodium hydroxide.

In other examples, the hydrolysis in step (a) is performed at elevated temperatures in the range of about 50°C to about 90°C, for example, temperatures in the range of about 60°C to about 90°C, about 70°C to about 90°C, about 50°C to about 80°C, and about 50°C to about 75°C, including each value within the specified ranges. Each possibility represents a separate example.

In yet other examples, the hydrolysis in step (a) is performed for a period of time in the range of about 1 to about 24 hours, including each value within the specified range. In other examples, the hydrolysis in step (a) is performed for a period of time in the range of about 1 to about 12 hours, including each value within the specified range.

In certain examples, the base is in excess of the PLA waste. In alternative examples, the PLA waste is in excess of the base.

In further examples, step (b) is performed and the acid is selected from hydrochloric acid, hydrobromic acid, phosphoric acid, sulfuric acid, and combinations thereof. Each possibility represents a separate example. In one example, step (b) is performed and the acid is sulfuric acid.

In various examples, step (b) is performed and removing unhydrolyzed PLA waste comprises solid-liquid separation.

In additional examples, the magnesium salt in step (c) is added in solid form. In alternative examples, the magnesium salt in step (c) is added as an aqueous solution. In further examples, the magnesium salt in step (c) is gradually added. In particular examples, the magnesium salt in step (c) is magnesium sulfate.

In additional examples, the obtained magnesium L-lactate salt is further subjected to subsequent purification. In other examples, the magnesium L-lactate salt is combined with magnesium L-lactate salt derived from fermentation of organic waste followed by subsequent purification.

Other objects, features and advantages of the present invention will become clear from the following description, examples and drawings.