

Application Of Glucono-Delta-Lactone Acid (GDL) In foods System: A Review

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Abstract

Glucono delta-lactone (GDL) is considered a processed product or ingredient being non-synthetic utilized in processing as organic for all applications safely. Mostly, food GDL uses relies on its chelating and acidic characteristics. GDL effects were inspected on cooked rice and the system of noodles. GDL is of many substances physical and chemical characteristics such as melting and boiling points, malleability, conductivity and capacity of heat besides functional characteristics where these characteristics collectively distinguished it and render it's as applications for food being safe. Glucose naturally exists in several foods. Once it reacts with atmospheric O₂, some oxidize to GA with the aid of glucose oxidase. Once GA takes place in H₂O presence, some of it might cyclize to GDL along with the involvement of the gluconate shunt where GDL is made from the GA aqueous solution through direct crystallization of GA for food utilization that might be produced in one of the 3 diverse methods (chemical, enzymatic and microbial). GDL food applications were performed in different products such as; dairy as a coagulant and gelling agent, bakery, cereal seed, meat, seafood, tofu, sauces and dressings. Moreover, GDL is of antioxidant activity, since it is occurring naturally as polyhydroxy acid (PHA) along with the metal chelating, antioxidant and moisturizing activity. Also, GDL considered an antimicrobial, extending shelf-life and minimizing total counts of microbes in products due to pH lowering which prevents bacterial growth and hastens the drying. Aims The aim of this review is to (1) add the GDL into food products such dairy products, meat products, beverages and ect due to its ability to reduce the pH value and create a gelatinous network; (2) study the capability of GDL to work as antioxidants and antimicrobial agents.

Keywords: Glucono-delta-lactone (GDL), Acid-induced-gelation

Introduction

Currently, glucono delta-lactone (GDL) is permitted under the program regulations of national organic at 7CFR 205.605(a). It is permitted as processed products or ingredient being non-synthetic that is labeled as "organic". Generally, it might be utilized in processing as organic for all applications safely. GDL might be added to foods as a pickling agent or curing, an agent of leavening, an agent to control pH, and as a sequestrant (21CFR 184.1318). It was petitioned originally for utilizing as a coagulant for tofu. Mostly, food GDL uses relies on its chelating and acidic characteristics. Initially, the taste is sweet when H₂O is added to

GDL. Then, to gluconic acid (GA), it hydrolyzes which offering a marginally sweet, somewhat acidic taste. In various foods, it is favored to other acids which provide robust, extratastes being acidic. If GDL is dissolved in H₂O, it will yield GA. Its slow attribute dissolution renders its taste fewer tart compared to other organic acids (Jungbunzlauer, 2008; Rubico and McDaniel, 1992).

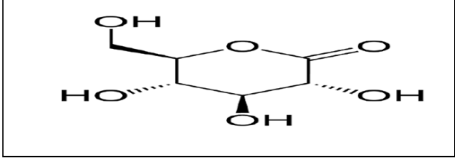
GDL has been utilized as an agent of coagulating in tofu processing as well as for the formation of curd cheese and milk heat stability in industries of dairy. GDL effects were inspected on cooked rice and noticed that the textural and microbial cooked rice qualities that were prepared with GDL and CH₃COOH at a concentration above 0.1 % exposed a noteworthy improvement. The cooked rice hardness that was prepared with both acidulants enlarged, signifying retrogradation took place quicker with acids presence. The study outcome may be beneficial in predicting rice-based noodles' textural qualities. Alternatively, the GDL utilized in the system of noodles was reported in Japanese udon. Such is a product of wheat and in the supermarket is currently marketed as a product as shelf-stable. GDL is added into a mix of udon flour for providing a low system of pH for prolonging such wheat-based noodles shelf life (Sumitra et al., 2006; Low et al., 2020).

Substances physical and chemical characteristics

Several substances physical characteristics differ based on the substance quantity whereas other substances physical characteristics do not differ based on the substance quantity, the volume and mass do not differ based on the substance quantity but, melting and boiling points, malleability, conductivity and capacity of heat do differ based on the substance quantity.

Table (1): General characteristics of Glucono-Delta- Lactone

Glucono-Delta- Lactone	Properties
Chemical formula	C ₆ H ₁₀ O ₆
Color	White, odorless practically, crystalline powder
CAS registry #	[90-80-2]
Trade name(s)	GDL
Chemical name	D-Glucono-1, 5-lactone; Gluconolactone; Delta-gluconolactone; 1, 5-Gluconolactone; D-(GA) lactone; GDL; D-(GA).

Food additives E numbers(other codes)	E575
Nat. Volatiles & Essent. Oils, 2021; 8(4): 11459-11474 pKa	3.70
pH	3.6
Solubility	Highly soluble, 5.9 x 10 ⁵ mg/L 25 °C sparingly soluble in alcohol
Density	1.68 g/ml
Vapor pressure	2.41 x 10 ⁻⁹ hPa at 25°C
Melting point	153 °C
Boiling point	398.5°C
Relative molecular mass	178.14 g/mol
Nature	Non-volatile organic acid, nontoxic
Representative structure	

(Kim et al., 2016;U.S.EPA 2019)

Functional characteristics of Glucono-Delta- Lactone

- 1- Leavening agent.
- 2- Bufferin agent.
- 3- Gelling agents.
- 4- Emulsification agent.
- 5- Pickling agent.
- 6- Chelating agent.
- 7- Edible coating
- 8- Color stabilizing.
- 9- H₂O-biding capacity.
- 10- Viscosity.

(Anonymous,1981: Liet al., 2011; Liu et al.,2019; Anonymous, 2020)

Antioxidant activity of GDL

GDL is occurring naturally as polyhydroxy acid (PHA) along with the metal chelating, antioxidant and moisturizing activity. GDL can be formed via D-glucose oxidation as enzymatic oxidation. Its capacity in scavenging free radicals explains its antioxidant activity (Anonymous, 1981; Zeng et al., 2017; Anonymous, 2020).

Antimicrobial activity of GDL

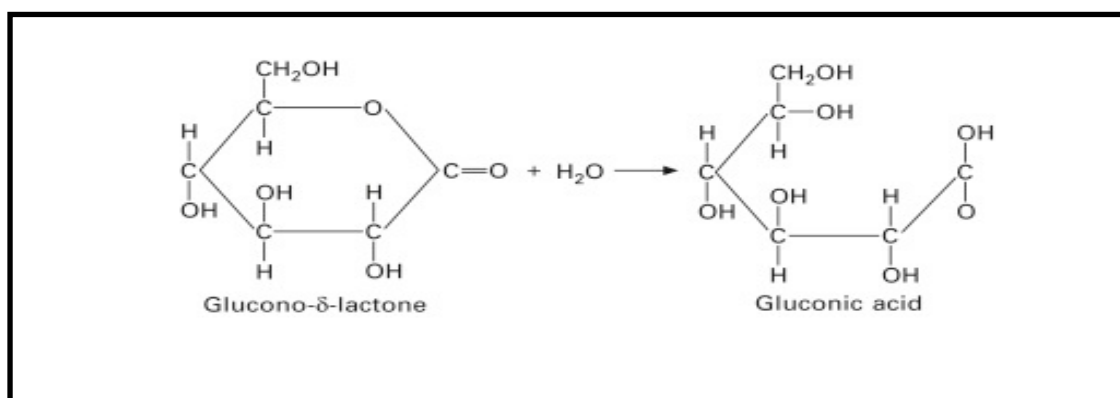
GDL is considered an antimicrobial, extending bologna shelf-life and minimizing total counts of microbes in products of pork and beef. The pH lowering prevents bacterial growth and hastens the drying (Bertelsen et al., 1995; Søltoft-Jensen and Hansen, 2005). GDL is found to be effective in preventing the growth of *Listeria monocytogenes* when used in combination with lactate in emulsion type meat products. Also, it was found that the lowering of pH by GDL together with lactate significantly improved the oxidative stability of the meat product and resulted in higher (Juncheret et al., 2000; Sameliset et al., 2002)

Substance source

Glucose naturally exists in several foods. Once it reacts with atmospheric O_2 , some oxidize to GA. GA can be present up to 1% in honey, up to 0.25% in wine, meat, rice, and vinegar. In honey, the amounts are higher due to the glucose oxidase enzyme presence, where salivary secretion occurs; hence, some enzymatic conversion is there. Wherever GA takes place in H_2O presence, some of it might cyclize to GDL (Ramachandran et al., 2006; Wong et al., 2008).

GDL composition

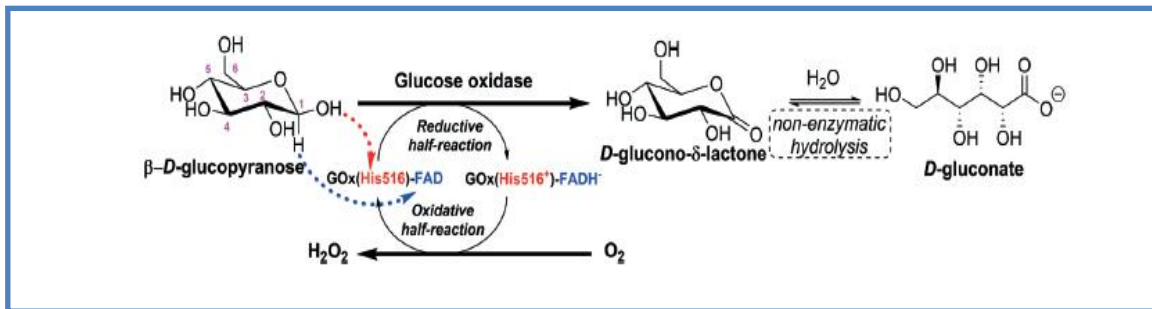
D-glucose is considered as aldohexose, it is a sugar of 6-C comprising the group as an aldehyde. If the group of aldehyde is oxidized to a carboxylic acid, D-GA is produced. The C adjacent to the acid group (C2) is called α (C2), the following one is β (C3), the following is γ (C4), and the following is the δ (C5). D-GA can react along with itself, producing lactone as a cyclic ester. If the reaction is between C1 of the acid group and δ (C5) of the hydroxyl group, the lactone is termed D-glucono- δ -lactone, D-glucono-1,5-lactone or D-36 GDL. GDL is considered as an inner, neutral GA cyclic ester formed via glucose acid fermentation. It takes place naturally in wine, honey, fruit juices and several products being fermented. GDL is a glucose derivative such as a saccharic acid and is a ring-shaped molecule. GDL exhibits 6 C atoms, and the group as OH is linked on each C atom (Ramachandran et al., 2006; Feiner, 2016).



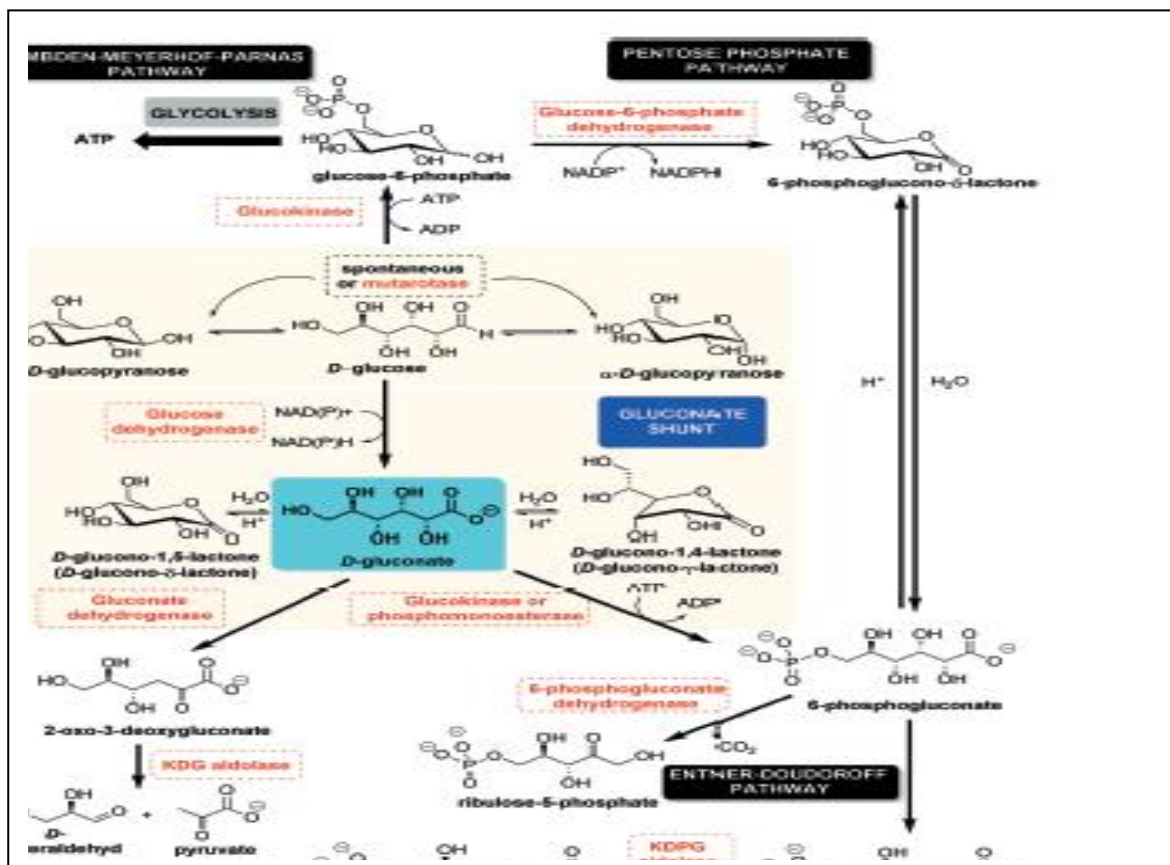
Figure(1):Glucono-delta -lactone (GDL) and(GA). (Feiner,2016)

Glucose oxidase: source and action mechanism

GA (penta-hydroxycaproic acid, Figure2) is formed from glucose via a simple reaction of dehydrogenation catalyzed via glucose oxidase. Aldehyde group oxidation on C-1 β -D-glucose to a carboxyl group causes glucono-delta-lactone (C₆H₁₀O₆) production and H₂O₂ (Figure 2). Glucono-d-lactone is extrahydrolyzed to GA either spontaneously or via lactone hydrolyzing enzyme, whereas H₂O₂ is decomposed to H₂O and O₂ via peroxidase enzyme (Ramachandran et al., 2006; Korneck et al., 2020).



Figure(2): Glucose oxidase mechanism in glucose oxidation to D-glucono-delta-lactone



(Leskovac et al., 2005; Bankar et al., 2009)

Figure (3): The gluconate shunt (Korneck et al., 2020)

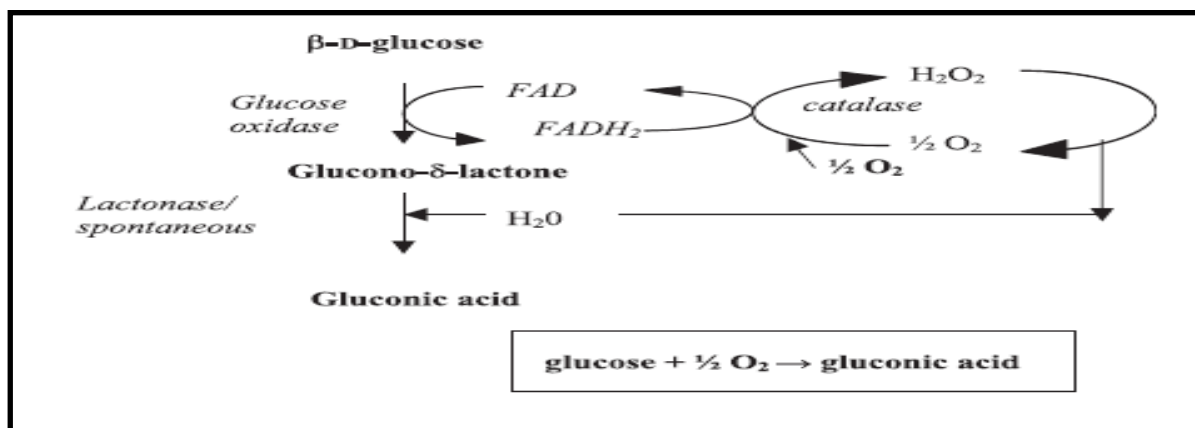
GDL production method

GDL is manufactured commercially from sources being renewable as carbohydrate via fermentation as microbial, then via processing of downstream. Throughout such a process, GDL is formed beside GA through the fermentation of glucose. GDL is made from the GA aqueous solution through direct crystallization of GA for food utilization in America that might be produced in one of the 3 diverse methods (FDA, 21CFR184.1318).

Table(2): Different acid glucono-delta-Lactone production methods

Methods	Oxidation	Used substances
Chemical	D-glucose oxidation	Bromine H ₂ O
Microbial	D-glucose oxidation	Microorganisms that are non-toxicogenic and non-pathogenic
Enzymatic	D-glucose oxidation	Enzymes resulting from such microorganisms

GDL is GA neutral cyclic ester. The acid is produced by aerobic carbohydrate source fermentation i.e., syrup of glucose derived from maize. Following fermentation, purification and crystallization will be performed. GDL dissolves quickly once it is added to the solution being aqueous. Then, it progressively hydrolyses to GA and the taste changes to slightly acidic from somewhat sweet (Haynes, 2010). GA is produced customarily via fermentation utilizing *Aspergillus niger*. Glucose catalase and oxidase genes from *A. niger* have been identified through isolation. GA formation might be improved by cloned genes utilization and alternative systems of the economic host may be developed (Nagarajan, 1992; BeMiller, 1992). Glucose oxidase-catalyzed D-glucose oxidation is a utilized method in commercial GDL production (Figure 3).



Figure(4): Fermentation method in glucose oxidation by *Aspergillus niger*(Ramachandran et al.,2006).

GDL application in the food system

Applications of food are occurring naturally in honey, fruit, wine, and kombucha tea that able to be formed via renewable raw materials fermentation which is readily bio-degradable, sustainable, and the products being safe. The food applications for the product group contain GA, GDL is a food additive as naturally-occurring is utilized as an acidifier, a sequestrant, or a curing, leavening agent or pickling. It is D-GA cyclic ester (Anonymous, 2020). In the food industry, GA is an organic acid mild that having applications. As indicated above, it is constituent being natural in honey and fruit juices and is utilized in foods pickling. Its glucono-d-lactone inner ester, divulges a sweet taste initially that becomes later somewhat acidic.

It is utilized in products of dairy and meat, principally in baked goods as a leavening agent component of products that pre-leavened. It is utilized as an agent of flavoring (i.e., in sherbets) and in decreasing fat absorption in cones and doughnuts. Food-stuffs comprising D-glucono-delta-lactone include yogurt, bean curd, bread, cottage cheese, meat and confectionery. In general, GA along with salts is utilized in food formulation (Table 3). Also, GA is utilized as a mineral (Ramachandran et al., 2006; Liu et al., 2019).

Table(3): Levels of (GDL) addition to foods (Anonymous, 1981)

Food type	Weighted mean%
Baked goods, baking mixes	0.31
Milk products	1.01
Cheese	1.00
Beverages	0.25
Meat products	0.15
Dairy products analogs	0.20

Table (4): Overview of major GDL food applications

Produce	Function	References
Dairy	Acidifier, coagulant, In the industry of dairy for the formation of cheese curd and for improvement of heat milk stability replacement of starter cultures	Ramachandran et al.(2006)
Bakery	Leavening agent	Jungbunzlauer (2009)
Cereal seed	wheat-based noodles, the quality of the noodles was improved dramatically	Low et al.(2020)
Meat	Curing agent, Phosphate replacement, stabilizer Slow acting acidulant in the processing of meat i.e., sausages	Yilmaz & Zorba (2010); Yimet et al.(2015)
Seafood	Replacement for sulfites, acidifier, preservation	Senturket et al.(2018)
Tofu	Soybean protein coagulation in tofu manufacturing	Chang(2006)
Sauces and dressings	A chelating agent, acidifier, preservation	PMP(2004)
Convenience food	Preservation, lowering pH	Low et al.(2020)
Microencapsulation	The stability of enhanced free cell and encapsulated of probiotic bacteria Lactobacillus casei and the increase in the efficiency of encapsulated	Nag(2011)
Edible Films and Coatings for Food Packaging Applications	reducing dehydration (as sacrificial moisture agent), controlling respiration, enhancing product appearance, improving mechanical properties, shelf-life	Senturket et al.(2018); Zhou et al.(2020)
Canning food	Texture and color loss prevention from excessive heat processing, browning inhibitor	Yilmaz & Zorba(2010)

1-Application of GDL in dairy products

A- A coagulant

The induced milk proteingelation via GDL is of significant standing in the acidified products milk processing. GDL utilize for casein micelles coagulation has been reported previously; nevertheless, no preceding studies have performed as proteomic analysis for investigating individual milk proteins coagulation. GDL effects on individual caseins coagulation and why proteins are considered as contribution being a novel. Several processes of production in the industry of dairy need a slow pH decline throughout manufacturing. In comparison to acidification microbiologically, GDL might be applied to milk at nearly any temperature and it permits excellent pH reduction reproducibility and control where various dairy processes need a slow reduction of pH. GDL is able to be utilized by replacing lactic acid bacteria in cottage cheese production, feta mozzarella and cheese (Rankin et al., 2006). Acidification being direct to form curding is an easy process for controlling compared to bacterial culture production. Also, the production of cheese is faster. Nevertheless, the cultured product of cottage cheese has a better texture and flavor (Makhalet al., 2013).

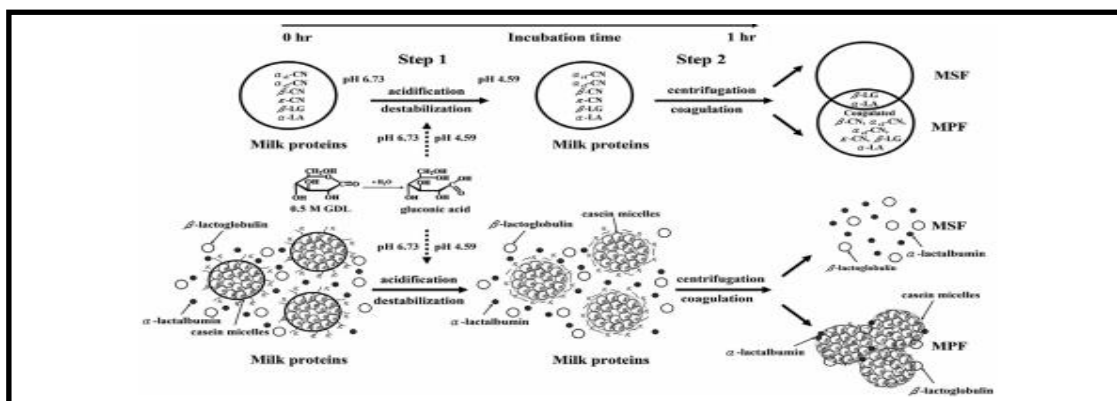


Figure (5): Reaction scheme for GDL effect on the milk proteins coagulation

(Chen et al., 2016)

B- Gelling agent

The acid-induced gelation of Buffalo milk (BM) using GDL was monitored using thromboelastography that can separate gelation into two phases, the onset gelation time and the time to get it firm. The pH at GT ranged from 5.5 to 5.9, which was higher than that reported for cow milk (CM) (pH 5.1–5). The pH at GT of BM increases with an increase in protein content, which may explain the higher pH at GT of BM as compared with CM. Also, the pH of BM at K20 was 5.40–5.65, which was higher than that of CM. Linear relations were found between GT, K20 of BM, and GDL concentration and gelation temperature (Khedka, 2016).

Acid-induced gels made from emulsions with higher protein concentrations had shorter gelation times but higher storage moduli, (G' and G'') fracture stresses and strains than gels formed by emulsions with lower protein concentrations. Increasing gelation temperature decreased the gelation time, fracture stresses and strains (Li et al., 2011). GDL is a commonly used as an acidifier. There has also been considerable research about the preparation of

high internal phase emulsions (HIPEs) with various proteins, including gliadin gelation using GDL. The HIPEs developed may be useful for encapsulating and delivering bioactive components, such as vitamins or nutraceuticals, through a variety of formats (Zeng, et al., 2017; Liu et al., 2019).

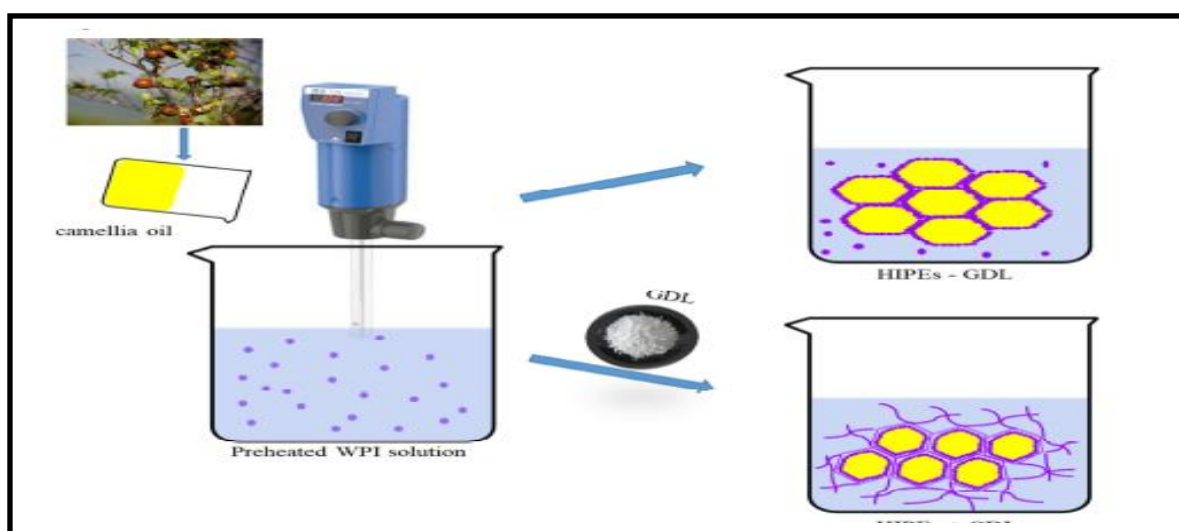


Figure (6): the presence and absence of GDL (Liu et al., 2019)

2- Application of GDL in Bakery leavening agent

GDL has been utilized for many years in high baking quality mixes for home utilization. GDL stability in dry mixes of a bakery might be enlarged and the final baked product quality improved via dry GDL coating, particularly in muffins mixes, bread, cakes, pizza, etc. Simultaneously, the fine-tuning of CO_2 release rate probability via temperature controlling permitted the GDL incorporation in products of high-tech canned refrigerated dough. Once freezing is not affecting its rising capacity, GDL has also been utilized already for decades in deep-frozen self-rising crust pizzas. In recent years, biscuits, finished cakes, and cookies besides cereal bars with GDL have been displayed on the market. GDL is utilized in bread as an agent of leavening. The produced GA once H_2O is added reacts with NaHCO_3 to form CO_2 gas, resulting in the dough rising. In comparison to other agents of leavening, it is of a slow to intermediate CO_2 rate release (Jungbunzlauer, 2008).

GDL is frequently added to mixes of cake due to having shelf-life being long, and it is often utilized in pastries. Up to 40% extra GDL might be added more as if is requisite for the reaction with NaHCO_3 . Such acidify the product and prevent mold growth (Feldberg, 1959). GDL slowly hydrolyzed to GA that then reacts with the NaHCO_3 to release CO_2 (Figure 6). Predominantly interesting is the hydrolysis of GDL slowness at room temperature and below, and its hastening with temperature surge: time of hydrolysis minimized to half for CO_2 every 10°C temperature raises. Thus, slight acid is produced throughout the preparation of dough and refrigerated storage with the results of a small loss of CO_2 at such stages (Singer, 2009).

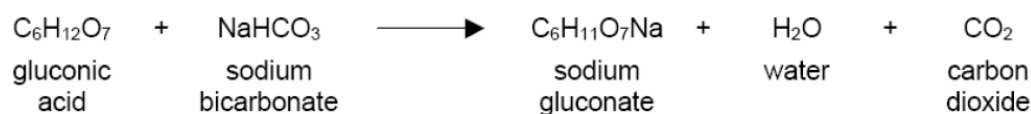


Figure (7): Leavening via GA reaction with NaHCO_3

(Singer, 2009)

When added into an aqueous solution GDL dissolves rapidly into the medium. Subsequently, it hydrolyses slowly to GA; thus, decreasing the pH progressively and continuously to equilibrium.

3- Application of GDL in Convenience food

The addition of GDL to a mix of udon flour was applied for providing a system of low pH for prolonging the shelf-life of such wheat-based noodles (Sumitra et al., 2006). Nevertheless, the direct acid addition into the system of flour may not be appropriate for noodles as rice-based; noodles of rice have a short shelf-life because of retrogradation of starch and microbial risk. Thus, it is mutual that noodles of fresh rice might just be made obtainable in certain parts close to the manufacturers. Through fresh rice noodles dipping in an acidic GDL bath, then through pasteurization in-pack, the quality of the noodles was improved dramatically. The treatment has delayed effectively the onset of long term fresh rice noodles retrogradation. Such discoveries carry excessive benefits to the manufacturers as the distribution of fresh rice noodles network may be extended to both existing and new customers (Low et al., 2020).

4- Application of GDL in Meat

GDL addition to processed meat frankfurters control the pH and encourage the ripening and curing process. It permits reduction up to 30% in the NO_2^- quantity added, and a residual NO_2^- reduction of 75% (Jungbunzlauer, 2008). Nevertheless, if ascorbic acid and GDL were utilized to replace a few NO_2^- in sausages

Turkish fermented, the product had lower production and poorer flavor. Mostly, cooked sausages are cured for developing the characteristics of red color. GDL stimulates the NO_2^- reaction with hemoglobin of meat at a level of little dosage as 0.1 to 0.2%, creating the stable cured meat products red color characteristic. Through lowering the pH utilizing GDL, the nitrous acid formation is hastened (Yilmaz and Zorba, 2010; Yimet al., 2015)

5- Application of GDL in Tofu coagulant

The soft-tofu made using GDL in Japan and acetic acid tofu made using acetic acid in Indonesia is mainly that the pH-induced precipitation of soy protein. Acidic whey tofu is considered to be more organoleptically superior to similar commercial products in China because of its special flavor, lightly sweet taste and good texture properties (Watanabe et al., 1997; Qiao et al., 2010). Soft tofu is frequently utilized in soups. It has an average of 84% as higher H_2O content compared to tofu as drier hard. Soft tofu is manufactured via coagulants i.e., nigari, CaCl_2 , or CaSO_4 , and through temperatures adjusting and rates of stirring to make a softer product. H_2O is either out of the pressed curds, or unpressed curds are left (Chang, 2006).

6- Application of GDL in Seafood

GDL aids in enzymatic browning inhibition in seafood through chelating metal ions that are requisite for their activation. It might thus replace SO_4 as an antioxidant. Combining GDL with SO_4 in the pre-freezing frozen shrimp's dip or in the blanching canned shrimp's brine renders it probable to substitute EDTA and minimize the level of sulfite by 50-90%. Also, GDL is utilized in the canned shrimp's canning brine both as a sorbate and mild acidulant and benzoate preservative enhancer agent. Moreover, it permits canning brine salt level reduction by 60-80% with no shrimp firmness loss. Therefore, the shrimps are in good health and have a better clean shrimp flavor, closer to the fresh product (Jungbunzlauer, 2008).

7- Application of GDL in Sauces and dressings

GDL is utilized to help and acidify preserve dressings of processed salad. Its chelating characteristics aid protection versus rancidity of oil through removing which catalyze fats oxidation (PMP, 2004).

8- Application of GDL in Beverages

Due to their unique characteristics, GDL, GA, and sodium gluconate are the perfect optimal as potent taste improvers for the industry of beverage. Comparing to the intensive standard acids taste i.e., malic, citric, and lactic acid, GA being of a slightly sweet, taste of mild acid with an obstinate effect on the tongue. GDL, the GA dry form is available also. Its taste as mild acid harmonizes very well with cola, ice tea, exotic and citric fruit aromas.

Sodium gluconate is can minimize the high-intensity sweeteners bitterness such as stevia besides caffeine and minerals. Moreover, it able to mask off-notes or artificial intense sweeteners aftertaste such as aspartame and saccharin, allowing them to be utilized widely in low- and mid-calorie soft, energy or sports drinks. The unique sodium gluconate characteristics qualify it as an alternative select for better and healthier tasting beverages. Drinks as soft made by simple ingredients blending, and hence with high throughput, the pH stabilization can take too long if utilizing GDL (Singer, 2011).

Conclusion

The study showed positive benefits of using GDL in molecular manufacturing. For example, it has been used in the dairy products to build a gelatinous network and then coagulants, in addition, improving the quality of baked products because GDL is considered as a fluffy agent. It also showed that GDL can successfully prevent the growth of microorganisms and thus prolonging the shelf life of food. The studies also showed its ability to improve the characteristics of other food products such as sauces, seafood, salads and drinks.

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