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Acid Hydrolysis and Optimization Techniques for Nanoparticles Preparation: Current Review

Zakiah Harun¹ · Agus Arsad¹ · Ai Ling Pang¹ · Mohd Abbas Ahmad Zaini² · Muslim Abdurrahman³ · Nuha Awang⁴ · Radzuan Junin⁵ · Rahmat Mohsin¹

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Abstract

Nanostarch is unique in that it is highly soluble, thermally stable, non-toxic and inexpensive. Hence, it is utilized in numerous well-established applications, including drug delivery, cosmetics, textiles, foods, and enhanced oil recovery (EOR). These applications take advantage of the special functions that can be achieved through modifications to the structure and properties of native starch. The most common method for the preparation of nanostarch with a relatively higher crystallinity and stability is acid hydrolysis. Technically, the properties of nanostarch are highly dependent on several factors during the hydrolysis process, such as the acid, concentration of acid, reaction time, reaction temperature, and source of starch. The production of nanostarch with desired properties requires a detailed understanding on each of the factors as they are inevitably affected the physical and chemical properties of nanostarch. Hence, it is vital to incorporate optimization technique into the production process to achieve the full potential of nanostarch. Therefore, the current review comprehensively elaborates on the factors that affect acid hydrolysis as well as the optimization techniques used in the preparation of nanostarch.

Keywords Acid hydrolysis · Optimization · Nanostarch · Acid concentration · RSM

✉ Agus Arsad
agus@utm.my

¹ UTM-MPRC Institute for Oil and Gas, School of Chemical and Energy Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, Skudai, 81310 Johor Bahru, Malaysia

² Centre of Lipids Engineering and Applied Research (CLEAR), Universiti Teknologi Malaysia, Skudai, 81310 Johor Bahru, Malaysia

³ Fakultas Teknik – Universitas Islam Riau, Jalan Kaharuddin Nasution, Workshop Gedung B, Lantai 2, Pekanbaru 28284, Indonesia

⁴ Plant Engineering Technology (PETech), Universiti Kuala Lumpur - Malaysian Institute of Industrial Technology (UniKL MITEC), Jalan Persiaran Sinaran Ilmu, Bandar Seri Alam, 81750 Masai, Johor, Malaysia

⁵ Department of Petroleum Engineering, School of Chemical and Energy Engineering, Faculty of Engineering, Universiti Teknologi Malaysia (UTM), 81310 Johor Bahru, Malaysia

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Introduction

Nanomaterials

The urgent demand for polymers from natural sources has contributed to the evolution and advancement of technologies, especially in nanomaterials. Nanomaterials are being widely used in a remarkable range of applications, including drugs delivery [1, 2], cosmetics [3, 4], textiles [5], foods [6, 7], and EOR [8–10]. Nanomaterials, ranging in size from 1 to 100 nm, are known for their small size and greater surface area per unit volume, which make them special and unique [11, 12]. However, the non-degradable properties of synthetically produced nanomaterials are a significant disadvantage, as it leads to their accumulation, which poses risks to the environment and health [13]. Therefore, non-toxic and biodegradable starch-based nanomaterials can serve as a viable alternative for synthetic material. For instance, nanomaterials derived from starch have been used extensively in bioactive ingredients and drugs encapsulation to improve the controlled release, stability, bioavailability, and bioactivity in drug delivery [14–17]. Besides that, the utilization of nanomaterials or nanoparticles in chemical EOR also has profoundly improved the rheological properties of fluids at various temperatures and salinities [8, 11]. Figure 1 illustrates the types of nanoparticles used in chemical EOR and their primary functions [13–15]. Additionally, the harmful disposal of synthetic plastic in the environment is able to be reduced by swapping to bioplastics incorporated with nanostarch, which have shown to be more compact film as the nanoparticles impede the formation of intermolecular hydrogen bonds between starch molecules [18].

The data in Fig. 2 shows the growth of research into the use of nanostarch, indicating that the starch-based nanoparticles are now widely recognised as an essential and well-studied development in various industries. Despite the extensive research into nanoparticles, as far as is known, no comprehensive review has been carried out on the extraction of nanostarch and the factors affecting the process conditions.

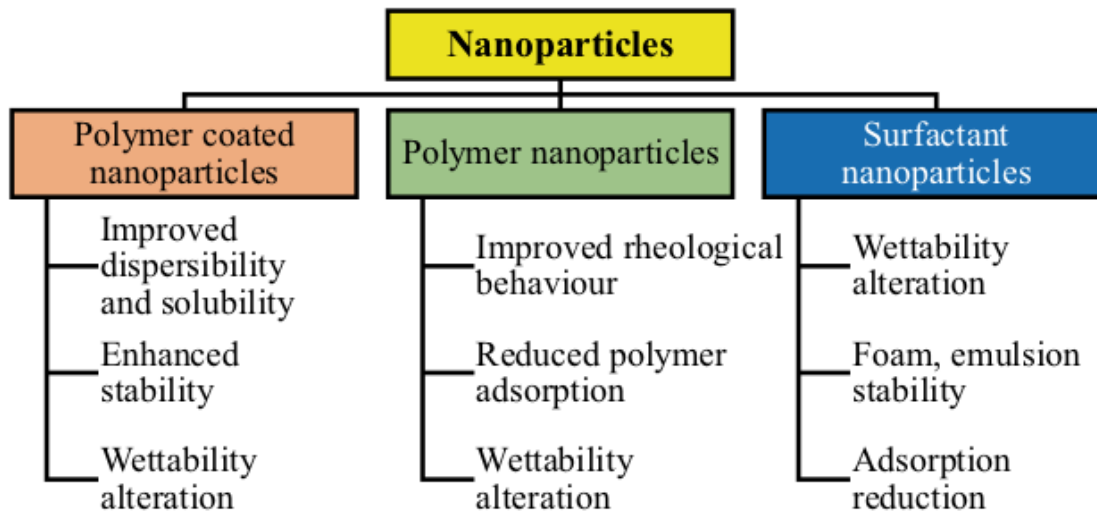


Fig. 1 Types of nanoparticles and their key function in chemical EOR

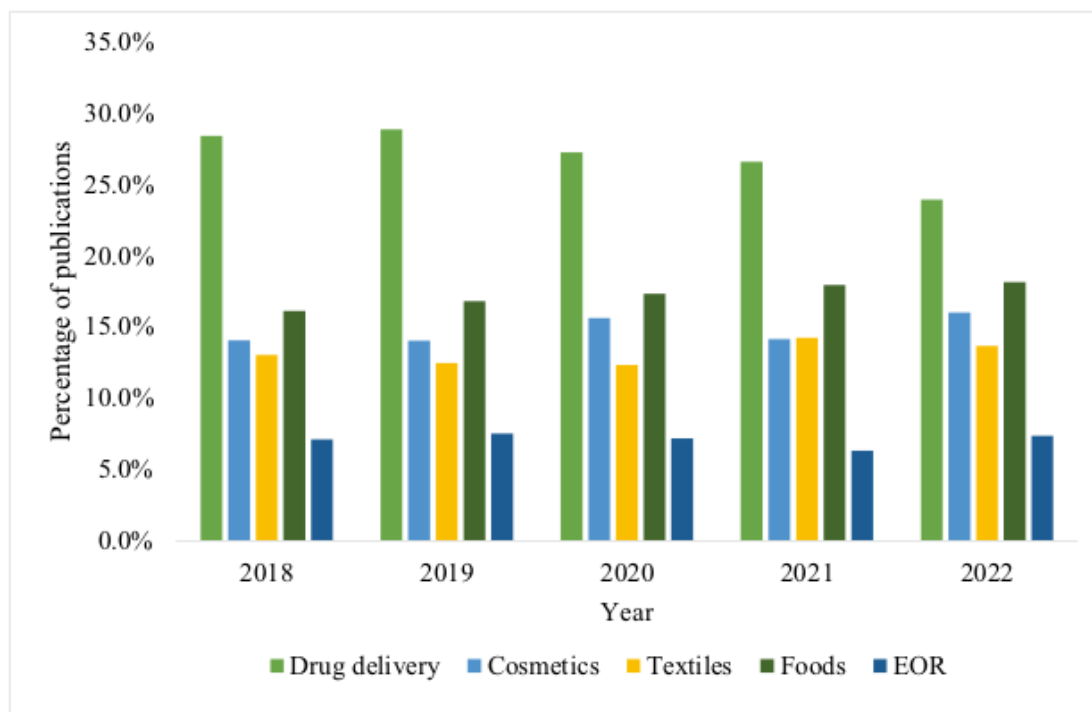


Fig. 2 Annual percentage of scientific publications related to nanostarch based on Elsevier data analysis as of 17th March 2022

Starch and Nanostarch

Starch is made up of complex microparticles, and is comprised of two main components, namely, amylopectin and amylose, with some water, lipids, phospholipids, soluble and insoluble fibers, as well as minerals [19]. Starch granules consist of concentric alternating amorphous and semi-crystalline polysaccharide molecules packed with amylopectin and amylose [20, 21]. In Fig. 3 (a), it can be seen that amylopectin is a highly branched α -D-(1-4)-glucan with α -D-(1-6)-glucan linkages at the branch point, whereas amylose, in Fig. 3 (b), is a linear chain composed of α -D-(1-4)-glucan units [22]. The ratio of amylopectin to amylose depends on the botanical origin and genetic background of each starch, but it usually consists of 70 to 80% of amylopectin and 20 to 25% of amylose [23]. This ratio varies according to the functional properties of the starch, such as its solubility, viscosity, gel stability, shear resistance, gelatinization, and retrogradation [24].

Starch is always preferred as a substitute for synthetic polymers because it is non-toxic, biodegradable, biocompatible, renewable, inexpensive, and widely available [18, 19, 25]. It can be used directly as native starch or treated starch. However, native starch is unsuitable for specific industrial applications due to several functional limitations, including low solubility, low retrogradation, restricted digestibility, poor thermal stability, and susceptibility to shear stress [22, 23, 26]. For this reason, starch is usually treated to diversify its structural and functional properties to fit various applications [27]. Generally, nanostarch is the most commonly used treated starch because it is known to have better physicochemical and biological properties, including higher solubility, greater reaction surface, absorptive capacity, and biological penetration rate, which are not available in conventional starch [28, 29].

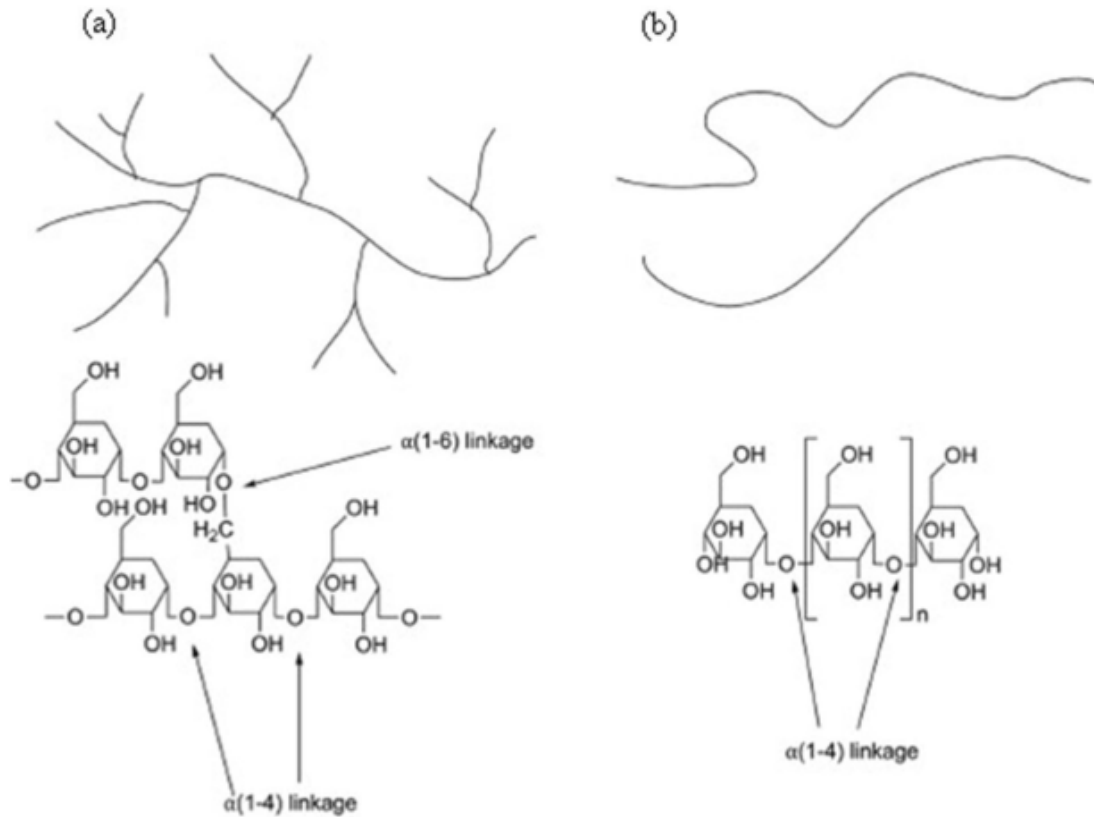


Fig. 3 Chemical structure of (a) amylopectin and (b) amylose [22]. Reproduced with permission from American Chemical Society

Table 1 presents the inherent properties of prepared nanostarches from different sources. Sharma et al. [32] exposed that kidney bean nanostarch gives higher solubility and swelling power compared to its native form. This advantage also agrees with Hung et al. [31] which have found that curcuma nanostarch can enhanced the starch solubility in hot water and cold water which is vital in producing good functional foods. Besides that, the dispersibility and thermal stability of the material has also shown to be improved by incorporating nanostarch-based carrier for loading bioactive compound in drugs delivery system [30, 36, 37, 39]. On top of that, Agi et al. [38] also proved that cassava nanostarch can potentially improve the oil recovery in EOR phase by generating higher viscosity during polymer flooding and eventually reduced the water and oil mobility ratio.

Preparation of Nanostarch

Starch can be chemically or physically altered to enhance or minimize unwanted features or to add new functional properties [27, 42]. A different approach to the modification of starch will lead to a different morphology, crystallinity, and size of the nanoparticles [32, 43, 44]. For example, media milling and ultrasound are used to physically modify the properties of starch. These methods are suitable for large-scale production and can provide long-term stability to the nanostarch without the assistance of a chemical stabilizer [30, 31]. However, both methods tend to destroy the crystalline structure of the starch due to the prolonged treatment time and additional energy required [36, 37]. Nevertheless, some studies have used ultrasound in combination with chemical modifications, such as

Table 1 Nanostarch from different sources and their inherent properties

| Starch | Particle size (nm) | Zeta potential (mV) | Relative crystallinity (%) | Water absorption capacity | Application | Remarks | References |
|----------------|--------------------|---------------------|----------------------------|---------------------------|--|--|------------|
| Quinoa | 166.25 | - | - | - | Drugs delivery | High storage stability | [30] |
| Curcuma | 194–455 | - | - | - | Functional foods | Increased solubility in cold water and hot water | [31] |
| Kidney bean | 257.7 | - | - | 54.76% | Food packaging | Enhanced solubility and swelling power higher | [32] |
| Taro | 130–360 | - | - | 14.03 g/g | Foods | Nanoparticles resulted in softer gel which suitable in food formulation | [33] |
| Corn | 40–60 | - | 43 | 9–13% | Nanofiller, encapsulation, bio-sensitive packaging | Crystallinity loss as acid concentration increased | [34] |
| Breadfruit | 145 | -17 | - | - | Food formulations, binders | Viscosity lower than native starch, loss of crystallinity | [35] |
| Broken-rice | 100–800 | - | - | 60–92% | Functional foods, pharmaceutical, cosmetics | Improved stability and dispersibility | [36] |
| Horse chestnut | 420.33 | 25.38 | 1.33 | 2.05–2.83 g/g | Food and drug formulations | Better thermal stability, increased viscosity, good antioxidant properties | [37] |
| Water chestnut | 535.21 | 41.29 | 5.56 | | | | |
| Lotus stem | 606.31 | 15.3 | 14.46 | | | | |
| Cassava | 538.2 | 24.1 | 23 | - | EOR | Increased crystallinity and viscosity, exhibit shear thinning behaviour | [38] |
| Potato | 42 | - | - | - | Food packaging, drugs delivery, barrier coating | Increased shelf-life and thermal properties | [39] |
| Water chestnut | 396 | - | - | - | Packaging material | Incorporate starch nanoparticles can increase thickness and burst strength for films development | [40] |
| Corn | 36–68 | - | 8 | - | Food packaging, drugs delivery | Thermal stability reduced | [41] |
| Cassava | 35–65 | | 0 | | | | |
| Yam | 8–32 | | 9 | | | | |

nanoprecipitation to (i) reduce the viscosity of the starch paste, thereby leading to a smaller particle size, and (ii) reduce the amount of precipitant [38, 39]. Besides that, enzymatic and acid hydrolysis are other examples of chemical modifications to native starch. These methods work similarly by introducing an enzyme or acid into the starch granules to cleave the glycosidic bonds [33–35]. The major drawback of enzymatic hydrolysis is that enzymes are highly selective, and can act only on specific chemical bonds [45]. Moreover, the rate of hydrolysis is also much slower than acid hydrolysis [46].

Acid hydrolysis is the preferred method for the preparation of nanostarch as it produces nanoparticles with a relatively higher crystallinity and stability compared to other methods [47]. Moreover, it is able to generate homogenous nanoparticles with the smallest size (5–7 nm) [48, 49]. In acid hydrolysis, starch is treated with an aqueous acid suspension at a temperature below its gelatinization point [24]. The hydroxonium ions will attack and dissociate the oxygen in the glycosidic bonds, thereby causing the links to break, and increasing the quantity of short linear chains, as shown in Fig. 4. The reaction occurs in two different phases, i.e., fast hydrolysis and slow hydrolysis. Fast hydrolysis involves the disruption of the starch granules in the amorphous region, resulting in the formation of small-sized nanoparticles, whereas in slow hydrolysis, the starch granules in the crystalline region are disrupted, leading to the formation of larger-sized nanoparticles [49, 50].

Factors Affecting Acid Hydrolysis

Several factors may affect the acid hydrolysis process and eventually lead to the formation of nanostarches with different physicochemical, technical, and mechanical properties. The acids, concentration of acid, reaction time, and reaction temperature are factors that significantly affect the process [51–54]. Moreover, factors, such as the source of starch and concentration of the starch, also have a stimulating effect on the disruption of the starch by acid hydrolysis [55]. Therefore, for the first time, this article provides a short concise review of the above-mentioned factors that influence the acid hydrolysis process.

Acid

During the hydrolysis process, the acid hydrolyses the amorphous starch granules on the surface before entering the densely packed crystalline inner region of the starch. Generally, the acid provides the hydrogen ions that react with the oxygen at the glycosidic bonds of the starch, and eventually, the links are hydrolysed [56]. Technically, hydrochloric acid (HCl) and sulfuric acid (H₂SO₄) are the two most commonly used inorganic acids in the hydrolysis process [22, 49, 57].

For instance, Saeng-on and Aht-Ong [58] conducted studies on acid hydrolysis using HCl and H₂SO₄ to produce banana and tapioca nanostarch. It was revealed that the use of H₂SO₄ was more effective in the hydrolysis of both nanostarches compared to HCl. Moreover, the highest crystallinities of about 47.13% and 60.06% were recorded for the H₂SO₄-hydrolysed banana and tapioca nanostarches, respectively. This was in agreement with the studies by Le Corre et al. [59] and Jiang et al. [60], who used H₂SO₄ on waxy maize starch for the isolation of nanostarch, and found a convincing increase in crystallinity (45–55%) and a stable nanostarch suspension ranging from –56 to –6.26 mV, respectively.

Additionally, Le Corre and Angellier-Coussy [49] conducted extensive studies on the influence of H₂SO₄ and HCl acid on hydrolysis process, where they obtained a lower yield

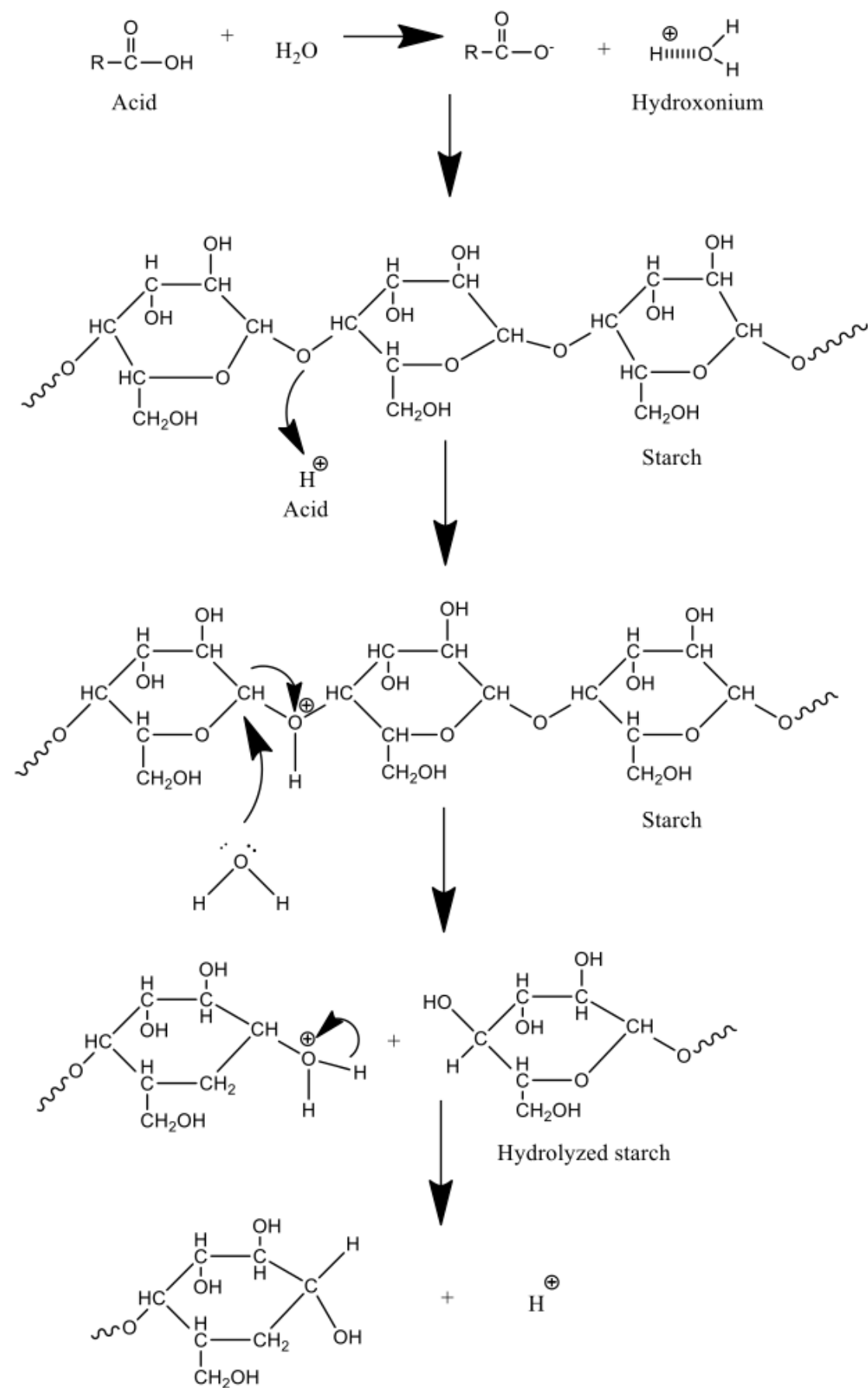


Fig. 4 Mechanism of acid hydrolysis on starch

of recovery with H_2SO_4 compared to HCl for the production of nanostarch. However, they convinced that the final suspensions were more stable with H_2SO_4 due to the presence of sulfate groups at the surface, as shown in Fig. 5. Unfortunately, even though the sulfate groups on the surface of the nanostarch aided in the dispersion and stability of the nanostarch in an aqueous solution, they also reduced the thermal stability of the nanostarch [61]. To address this issue, Yang et al. [62] suggested that 0.5 wt.% of ammonia be added to the H_2SO_4 nanostarch suspension prior to the subsequent centrifugation to improve the thermal stability of the nanostarch and make it suitable for chemical alterations.

Basically, a critical element in the preparation of nanostarch is to avoid destroying the crystalline structure of the starch granules while removing the entire amorphous portion on the surface of the granules. HCl and H_2SO_4 are categorized as strong acids, and therefore, they have a strong dissociation constant (HCl 1.3×10^6 , H_2SO_4 1.0×10^3), which can lead to excessive hydrolysis of the glycosidic bonds, including in the densely packed crystalline region of the starch granules [22, 49, 57]. This will cause the starch to lose its crystallinity as well as reduce the recovery yield of the nanostarch. Furthermore, Dufresne [63] also highlighted that nanostarches obtained by HCl and H_2SO_4 have a significant tendency to agglomerate, especially in dry powder form, thereby limiting their actual commercial applications.

Recently, organic acids have gradually attract more attention because of several advantages such as low corrosion of equipment, fully recyclable, environmental friendly, and introduction of functional groups which provided more stability in the starch suspension [38, 64]. For example, Agi et al. [38] used ascorbic acid from natural sources to isolate crystallized nanostarch from native cassava. When cassava is treated with ascorbic acid, a corresponding ester emerges on the surface of the nanostarch, and the remaining valences dissociate, resulting in electrostatic repulsions which contribute to their stability in water. This is due to the presence of randomly dispersed negatively charged ester groups on the surface of the nanostarch that enhance the zeta potential value as the hydrolysis time is prolonged [65]. Besides that, citric acid and stearic acid are also suitable to chemically modify the starch granules as they are obtained from natural sources and safe to be utilized especially in foods product. For example, Singh et al. [66] treated Moth bean (*Vigna aconitifolia*) using a combination of citric acid and stearic acid to produce resistance starch with higher crystallinity and solubility. Similar study was conducted by Cuthbert et al. [67] using stearic acid in isolation of tef and maize starch. They reported that the relative crystallinity for both nanostarches increased with time but no significance difference observed in its thermal stability. Organic acid also possesses appealing

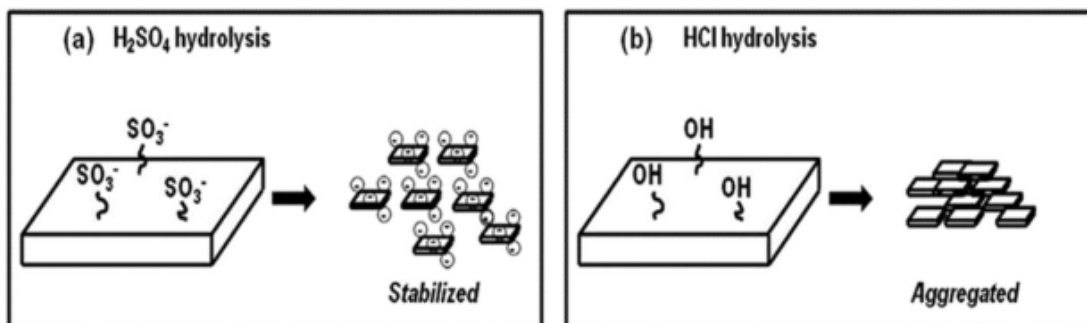


Fig. 5 Comparison between nanostarch prepared with (a) H_2SO_4 hydrolysis and (b) HCl hydrolysis [49]. Reproduced with permission from Elsevier

practical and chemical characteristics, such as recoverability and ease of handling and storage. Besides, it is highly selective and effective in the industrial production of modified-starch, environmentally friendly, as well as non-corrosive to the process equipment.

Concentration of Acid

Acid hydrolysis is also affected by the concentration of acid used in the preparation of nanostarch [33, 51, 52, 55, 58]. A higher acid concentration means a higher number of hydrogen ions will be released into the suspension, leading to more cleavage of glycosidic bonds in both the amorphous and crystalline regions of the starch [32, 36, 68]. Numerous studies have been conducted to evaluate the effect of various concentrations of acid on the preparation of nanostarch. For example, Jiang et al. [69] examined the properties of waxy nanostarch prepared in different concentrations (0.25 M, 0.5 M, 1.0 M, 1.5 M) of HCl. It was found that the nanostarch prepared with 1.5 M of HCl exhibited the smallest particle size (3.59×10^3 nm), whereas the one that was prepared with 0.25 M of HCl showed the largest particle size (2.85×10^4 nm). This was supported by Md Shahrodin et al. [70], who confirmed that a higher acid concentration (3.4 M of H_2SO_4) gave the smallest particle size (5 nm), whereas a low acid concentration (2.8 M of H_2SO_4) resulted in a larger particle size (20 nm). In addition, the results postulated that the crystallinity of the produced nanostarch initially increased from 24.48 to 25.96% before decreasing to 25.20% as the HCl concentration increased [69]. This pattern was due to the initial damage to the crystalline area after the amorphous region had been completely hydrolysed [42]. Additionally, Md Shahrodin et al. [70] also observed a trend of increase in the percentage of the recovery yield of cassava nanostarch as the concentration of H_2SO_4 increased. The highest yield reported was 1.1%, which was produced from 3.4 M of H_2SO_4 , whereas a concentration of 2.8 M of H_2SO_4 gave the lowest yield of 0.54%. However, Saeng-on and Aht-Ong [58] revealed a contrasting trend with regard to the effect of the acid concentration on the percentage yield of the nanostarch. They found that the nanostarch prepared with a higher acid concentration (4.5 M of H_2SO_4) gave a lower percentage yield. They explained that the corrosion and degradation that occurred at higher acid concentrations caused more cleavage of the starch chains, leading to the loss of the recovery yield.

Low recovery yield has been the ultimate drawback of using H_2SO_4 hydrolysis not only in the preparation of nanostarch, but also in the preparation of nanocellulose. Hamad and Hu [71] obtained low nanocellulose yields of around 35% since they could not reconcile the rapid change in nanocellulose yield at acid concentrations ranging from 56 wt.% to 58 wt.%. Whereas, Chen et al. [72] investigated the yield pattern of nanocellulose from bleached eucalyptus kraft dry lap pulp by varying the H_2SO_4 concentration between 50 and 64 wt.%. They noticed that 58 wt.% of H_2SO_4 is the sharp transition point where nanocellulose gives the highest yield (75.6%) with particle diameter of 204.6 nm, at a reaction temperature of 56 °C within 210 min. Subsequently, the residue yield of nanocellulose was rapidly decreased as the H_2SO_4 concentration increases beyond 58 wt.%.

Reaction Time

Another factor that has a significant influence on the process of acid hydrolysis is the reaction time [58, 60]. For instance, studies from Cuthbert et al. [67] that used stearic acid for tef and maize nanostarch isolation have reported a reduction in the percentage yield for both starch as the hydrolysis time increased. The percentage yield of tef and maize

nanostarch reduced from 75.4 to 30.2% and 74.4 to 29.8%, respectively after 10 min of reaction time. However, the particle size for both nanostarches was found to increase as the hydrolysis time increased. The relative crystallinity of the tef and maize nanostarch increased significantly with hydrolysis time from 18.3 to 53.7% and 26.7 to 53.3% after 10 min, respectively.

Chen et al. [72] discovered that there was a reduction in the particle size and percentage of recovery yield of nanocellulose with a prolonged hydrolysis time. The particle diameter and recovery yield were also found to be reduced from 147.6 to 130.5 nm and 51.1 to 28.0%, respectively after an extension of 90 min of hydrolysis time. Besides that, Saeng-on and Aht-Ong [58] noticed an increase in the percentage of crystallinity and yield of banana nanostarch, before gradually decreasing after a certain critical time. It was found that the hydrolysis of 3.5 M of H_2SO_4 on banana starch resulted in a remarkable improvement in the percentage of crystallinity from 18.95% at 3 h to 47.13% at 7 h, before reducing to 18.16% at 15 h. A similar trend was also shown in the percentage of yield, which initially increased from 52.97% at 3 h to 67.24% at 7 h, before reducing to 26.97% at 15 h. However, the size of the nanostarch particles was observed to decrease linearly from 3 h to 7 h. Initially, the native banana starch granules (2.9×10^4 nm) experienced a rapid hydrolysis reaction before entering into a slow rate of hydrolysis to produce nanostarch granules of between 30 and 70 nm after 7 h of hydrolysis with 3.5 M H_2SO_4 at 40 °C. Meanwhile, the hydrolysis of 3.0 M HCl on banana starch resulted in a significance reduction in the percentage of crystallinity and yield upon reaching 7 h of reaction time. The highest percentage of crystallinity reported was 40.15% with 83.02% of percentage yield at 5 h of reaction time.

In another example, Luzi et al. [73] studied the acid hydrolysis reaction time for the extraction of *Posidonia Oceanica* nanocellulose using a concentration of 64 wt.% of H_2SO_4 . The results showed that the nanocellulose prepared within 60 min had a smaller particle size (5–10 nm) than the nanocellulose prepared within 30 min (15–20 nm). They also revealed that the percentage of recovery yield increased from 8 to 15% with an additional 30 min of reaction time.

Reaction Temperature

Temperature is one of the most vital factors that affects the process of acid hydrolysis, particularly for polysaccharides [29, 53]. Generally, acid hydrolysis is carried out at a temperature below the gelatinization point of polysaccharides to avoid the (i) disruption of the crystalline structure of the starch, and (ii) reduction of the recovery yield [61]. There are several works that investigated the effects of reaction temperature on the acid hydrolysis of polysaccharides. For example, a study by Kapdan et al. [74] reported that there was an increase in the total sugar yield from 17.4 to 20.3 g/dm³ when the reaction temperature was increased from 90 to 130 °C, respectively at a constant hydrolysis time of 30 min in a dilute H_2SO_4 condition. However, the opposite trend was discovered in a study by Saeng-on and Aht-Ong [58] on the acid hydrolysis (HCl and H_2SO_4) of banana and tapioca starch. They concluded that a lower reaction temperature (40 °C) was more suitable than a higher reaction temperature (50 °C and 60 °C) for the highest crystallinity and yield of nanostarch. This is because the heat generated at 50 °C and 60 °C can cause the starch granules to corrode and disintegrate, resulting in a decrease in the degree of crystallinity and percentage of recovery yield. The crystallinity of the starch may be disrupted when the processing

temperature exceeds the starch gelatinization point, which will cause the starch to swell up, thereby reducing the percentage of recovery yield [61].

Meanwhile, Kanchanalai et al. [75] found that the concentration of xylose increased slowly (within 7 h) when the reaction temperature used was 80 °C. However, when the reaction temperature was increased to 90 °C, the xylose concentration was found to rapidly increase in the first 60 min before reaching its maximum value of between 28 and 29 g/L. Subsequently, the xylose concentration becomes constant. A similar trend was observed by Zhang et al. [76], who illustrated that as the temperature was increased from 120 to 130 °C, the yield of XOS increased sharply from 17.76 to 29.64%, before slowly decreasing as the temperature reached 140 °C. This finding indicated that the optimum temperature for the production of XOS with a maximum yield was 130 °C.

Source of Starch

Acid hydrolysis is also affected by the botanical origin of the material sources [55, 76]. The properties of nanostarch depend immensely on the source and geotype of the starch, which differ mainly according to the fraction and organization of its amylose and amylopectin [57]. Native starches are differentiated according to their X-ray diffraction (XRD) patterns, which denote them as A-, B-, or C-type starches [77]. A-type starches are mainly associated with cereal crops like maize and wheat, which have strong diffraction peaks at around $2\theta = 15^\circ, 17^\circ, 18^\circ,$ and 23° . B-type starches are often derived from plant tubers, such as potato and canna. The most substantial diffraction peak for B-type starches has been observed at $2\theta = 17^\circ$, with several other low peaks at $2\theta = 5.6^\circ, 20^\circ, 22^\circ,$ and 24° . Meanwhile, C-type starches are crystalline hybrids of A- and B-type starches, such as pea and bean starches [32, 65, 78]. Different types of starches demonstrate different hydrolysis mechanisms. For instance, B-type starches are less vulnerable and display a higher resistance to acid degradation than A- and C-type starches. This could be seen in the work by Xia et al. [52], in which it was reported that there was a slow initial hydrolysis rate in the B-type *Fritilla ussurensis* compared to the A-type maize starch and C-type Chinese yam. After hydrolysis with 2.2 M of HCl for 32 days, the highest recovery yield was observed in the *Fritilla ussurensis* (50.8%), followed by the Chinese yam (40%), and maize (35.8%). In addition, starches with random-shaped granules are more susceptible to acid than starches with smooth and regular shapes. This is because the pores in the random-shaped granules create channels for the hydrogen ions to penetrate and attack the amorphous region [79].

Perez Herrera et al. [80] investigated the influence of different starch morphologies on the properties of acid-treated A-type wheat, oat, barley, and B-type potato starch. They noticed that all the types of nanostarch with comparable amylose levels exhibited significant differences in their particle size distribution, thermal stability, and rheological properties after being treated with 3.16 M of H₂SO₄ at 40 °C for 6 days. The diameter of the different nanostarch particles varied significantly, with the largest diameter being recorded in wheat (70–226 nm), followed by barley (62–167 nm), whereas the oat and potato starches exhibited the smallest particle size distribution of 55–130 nm and 49–155 nm, respectively. On top of that, starches of the same type, but obtained from different sources, also have different crystallinities after acid hydrolysis. For instance, Md Shahrodin et al. [70] found that the crystallinity of cassava starch changed from C-type to B-type after limerization. The A-type diffraction peaks, which were initially present in the native cassava, slowly disappeared, while the B-type peaks became sharper as the concentration of H₂SO₄ gradually increased. However, the results were not in line with the finding by Rolland-Sabaté et al.

[81], who reported that limerization increased the crystallinity of either the acid-thinned A- or B-type starch to that of the C-type. Meanwhile, Sivamani et al. [82] hydrolysed cassava starch using HCl and H₂SO₄, and discovered no apparent change in the crystallinity of the native starch, which was initially an A-type starch. However, the intensity of the diffraction peaks of the A-type starch had become higher, indicating that the crystallinity had increased. Table 2 presents a summary of the acid hydrolysis conditions from different studies.

Hydrolysis Optimization Techniques

Hypothetically, each type and level of factors are equally essential and significantly affect the acid treatment process. The production of nanoparticles with desired properties requires a detailed understanding of each of the parameters as they inevitably affect the production yield. A low yield of nanoparticles significantly contributes to a rise in the manufacturing cost, and therefore, maximizing the yield (assuming identical production procedures) can result in considerable cost savings. The most promising way to improve the production yield is through process optimization [34, 80]. Several optimization techniques have been extensively utilized in the acid hydrolysis process, including a statistical design of experiment (DoE), such as the central composite design (CCD) and Box-Behnken design (BBD), artificial neural networks (ANN), conventional experimental design, and orthogonal design [53, 69, 71, 72].

For instance, Dong et al. [85] used a three-factor CCD in response surface methodology (RSM) to determine the optimum reaction conditions for the preparation of nanocellulose from wood pulp by hydrolysis with H₂SO₄. They reported a maximum predicted yield of 66–69% at the optimum process conditions of 57–58 wt.% acid concentration, 64–67 °C reaction temperature, and 134–156 min of reaction time. In these conditions, the sulfate group density was predicted to be between 241 and 265 mmol/kg. Table 3 presents a summary of the optimization techniques used in acid hydrolysis with optimum reaction conditions. Zou et al. [87] isolated nanocellulose crystals by means of hydrolysis with H₂SO₄ by adapting the three-factor, three-level orthogonal design. The orthogonal test demonstrated that the optimum reaction conditions to obtain the highest yield (90.14%) were at reaction time of 108 min, reaction temperature of 43 °C, and H₂SO₄ concentration of 38 wt.%. The particle diameter was found to be in nanosize which is 20–30 nm.

Additionally, Saeng-on and Aht-Ong [58] adapted a conventional experimental method to find the optimum conditions for the preparation of nanostarch from banana and tapioca. The measurements were carried using Duncan's multiple range tests, while the analysis of variance (ANOVA) was used to compare the means. The studies found that the optimum conditions to produce nanostarch were to use 3.5 M of H₂SO₄ at 40 °C for 7 h and 10 h for banana and tapioca starch, respectively. These conditions resulted in a yield of 67.24%, with a crystallinity of 47.13% in the banana nanostarch, while the yield for the tapioca nanostarch was about 85.66%, with a crystallinity of 60.06%.

Technically, the conventional method employed by Saeng-on and Aht-Ong [58] is called a one-factor-at-a-time method, where an experiment is carried out by manipulating one factor at a time, while keeping the others constant. Optimizing an experimental process using this method is very inefficient as it is time-consuming and can increase the overall operating costs. Despite its simplicity and ease of handling, this method can only operate at its best when a small number of variables are involved [88]. However,

Table 2 Summary of acid hydrolysis condition from different studies

| Starting material | Acid | Acid conc. | Time | Temp. (°C) | Yield (%) | Particle size (nm) | Application | Reference |
|--------------------------------|--|-------------|--------|------------|-----------|----------------------|--------------------------------|-----------|
| Cassava starch | Ascorbic acid | – | 5 d | 60 | – | 6.97–200 | EOR | [38] |
| Bleached eucalyptus kraft pulp | H ₂ SO ₄ | 64 wt. % | 30 min | 45 | 16 | 181 (L) 7 (W) | – | [78] |
| | Oxalic acid/H ₂ SO ₄ | 50/10 wt. % | 1 h | 80 | 82 | 618 (L) 60 (W) | | |
| | | | 2 h | | 77 | 302 (L) 15 (W) | | |
| | | | 3 h | | 75 | 278 (L) 10 (W) | | |
| | | | 5 h | | 70 | 211 (L) 10 (W) | | |
| Wheat starch | H ₂ SO ₄ | 3.16 M | 7 d | 40 | – | 30–80 | Food processing | [47] |
| Waxy maize starch | HCl | 0.25 M | 3 h | 50 | – | 2.01×10 ⁴ | – | [69] |
| Waxy corn starch | | 0.5 M | | | | 9.46×10 ³ | | |
| | | 1.0 M | | | | 5.63×10 ³ | | |
| | | 1.5 M | | | | 3.59×10 ³ | | |
| Waxy rice starch | HCl | 2.2 M | 7 d | 35 | – | 20–420 | Resistant starch | [83] |
| | | | 10 d | | | 30–300 | | |
| Cassava peel | H ₂ SO ₄ | 3 M | 5 d | 37 | 9.8 | – | – | [82] |
| | HCl | 2.5 M | 7 d | | 7.6 | | | |
| Banana starch | HCl | 3.0 M | 5 h | 40 | 83.02 | 30–70 (L) | Filler for bio-based materials | [58] |
| | H ₂ SO ₄ | 3.5 M | 7 h | | 67.24 | | | |
| Tapioca starch | HCl | 3.0 M | 7 h | | 54.43 | | | |
| | H ₂ SO ₄ | 3.5 M | 10 h | | 85.66 | | | |

Table 2 (continued)

| Starting material | Acid | Acid conc. | Time | Temp. (°C) | Yield (%) | Particle size (nm) | Application | Reference |
|----------------------------------|--------------------------------|-------------|--------|------------|-----------|--------------------------|------------------|-----------|
| Waste xylan | Acetic acid | 20 wt. % | 20 min | 140 | 45.86 | – | XOS as prebiotic | [76] |
| | | 30 wt. % | 30 min | 120 | 17.76 | | | |
| | | 30 wt. % | 30 min | 130 | 29.64 | | | |
| | | 30 wt. % | 20 min | 130 | 32.19 | | | |
| Wheat starch | H ₂ SO ₄ | 3.16 M | 6 d | 40 | – | 70–226 | – | [80] |
| Barley starch | | | | | | 62–167 | | |
| Oat starch | | | | | | 55–130 | | |
| Potato starch | | | | | | 49–155 | | |
| Xylan | H ₂ SO ₄ | 10–50 wt. % | 7 h | 80–100 | – | – | – | [75] |
| <i>Posidonia oceanica</i> leaves | H ₂ SO ₄ | 64 wt. % | 30 min | 45 | 8 | 15–20 (W) 500–650 (L) | Nanocomposites | [73] |
| | | | 60 min | | 15 | 5–10 (W) 200–450 (L) | | |
| Waxy maize starch | H ₂ SO ₄ | 3.16 M | 5 d | 40 | – | 80–110 (L) 30–60 (W) | Films | [60] |
| Cassava starch | H ₂ SO ₄ | 2.8 M | 5 d | 37 | 0.54 | 5–20 (W) | – | [70] |
| | | 3.0 M | | | 0.60 | | | |
| | | 3.2 M | | | 0.73 | | | |
| | | 3.4 M | | | 1.10 | | | |

Table 2 (continued)

| Starting material | Acid | Acid conc. | Time | Temp. (°C) | Yield (%) | Particle size (nm) | Application | Reference |
|--|--------------------------------|--------------|---------|------------|-----------|----------------------|--|-----------|
| Bleached eucalyptus kraft dry lap pulp | H ₂ SO ₄ | 56 wt. % | 90 min | 70 | 30 | 281.2 (L) | Cellulose nanocomposites, smart windows, LED | [72] |
| | | 58 wt. % | 180 min | 56 | 71.7 | 228.2 (L) | | |
| | | 58 wt. % | 210 min | 56 | 75.6 | 204.6 (L) | | |
| | | 62 wt. % | 105 min | 40 | 64.8 | 177.1 (L) | | |
| | | 62 wt. % | 75 min | 50 | 70.3 | 143.0 (L) | | |
| | | 62 wt. % | 30 min | 60 | 71.6 | 189.3 (L) | | |
| | | 62 wt. % | 60 min | 60 | 69.4 | 149.0 (L) | | |
| | | 64 wt. % | 15 min | 45 | 51.1 | 147.6 (L) | | |
| | | 64 wt. % | 105 min | 45 | 28.0 | 130.5 (L) | | |
| | | Wheat starch | HCl | 0.36 N | 4 h | 40 | | |
| 0.36 N | 24 h | | | | | 2.29×10 ⁴ | | |
| 0.72 N | 4 h | | | | | 2.45×10 ⁴ | | |
| 0.72 N | 24 h | | | | | 2.27×10 ⁴ | | |
| 0.36 N | 4 h | | | | | 2.41×10 ⁴ | | |
| 0.36 N | 24 h | | | | | 2.25×10 ⁴ | | |
| 0.72 N | 4 h | | | | | 2.54×10 ⁴ | | |
| 0.72 N | 24 h | | | | | 2.25×10 ⁴ | | |

Table 2 (continued)

| Starting material | Acid | Acid conc. | Time | Temp. (°C) | Yield (%) | Particle size (nm) | Application | Reference |
|----------------------|--------------------------------|------------|------|------------|-----------|----------------------|----------------|-----------|
| Potato starch | HCl | 0.36 N | 4 h | 40 | – | 4.28×10 ⁴ | – | [57] |
| | | 0.36 N | 24 h | | | 4.13×10 ⁴ | | |
| | | 0.72 N | 4 h | | | 4.39×10 ⁴ | | |
| | | 0.72 N | 24 h | | | 4.23×10 ⁴ | | |
| | H ₂ SO ₄ | 0.36 N | 4 h | | | 4.43×10 ⁴ | | |
| | | 0.36 N | 24 h | | | 4.45×10 ⁴ | | |
| | | 0.72 N | 4 h | | | 4.45×10 ⁴ | | |
| | | 0.72 N | 24 h | | | 4.34×10 ⁴ | | |
| Pea starch | HCl | 0.36 N | 4 h | 40 | – | 2.64×10 ⁴ | – | [57] |
| | | 0.36 N | 24 h | | | 2.57×10 ⁴ | | |
| | | 0.72 N | 4 h | | | 2.63×10 ⁴ | | |
| | | 0.72 N | 24 h | | | 2.57×10 ⁴ | | |
| | H ₂ SO ₄ | 0.36 N | 4 h | | | 2.61×10 ⁴ | | |
| | | 0.36 N | 24 h | | | 3×10 ⁴ | | |
| | | 0.72 N | 4 h | | | 2.58×10 ⁴ | | |
| | | 0.72 N | 24 h | | | 2.56×10 ⁴ | | |
| Waxy maize starch | H ₂ SO ₄ | 3.16 M | 7 d | 40 | 5.05 | 200–300 | – | [84] |
| Maize starch | HCl | 2.2 M | 32 d | 35 | 35.8 | 9.4×10 ³ | – | [52] |
| <i>F. ussurensis</i> | | | | | 50.8 | 1.76×10 ⁴ | | |
| Chinese Yam | | | | | 40 | 1.53×10 ⁴ | | |
| Pea starch | H ₂ SO ₄ | 3.16 M | 5 d | 40 | – | – | Nanocomposites | [62] |

*L length, W width

Table 3 Optimization techniques used in acid hydrolysis

| Method | Starting material | Optimum condition | Optimum response | Remarks | Reference |
|-----------------|----------------------------|---|---------------------------|---|---|
| CCD-RSM | Pomegranate peels waste | Acid conc. (wt.%) Temp. (°C) Time (min) | 5 100 30 | Yield (%) 12.76 | Model fitted well with experimental data [86] |
| ANN, BBD-RSM | Waste potatoes | Acid conc. (wt.%) u/s time (min) <i>S. cerevisiae</i> (g/L) | 2.1 10.7 19.2 | Yield (g/L) 65.8 | Model fitted well with experimental data [45] |
| Experimental | Banana, tapioca | Acid conc. (M) Temp. (°C) Time (h) | 3.5 40 7-10 | Yield (%) 67.24, 85.66 | Reaction time affect the yield most [58] |
| CCD-RSM | Wood pulp | Acid conc. (wt.%) Temp. (°C) Time (min) | 57-58 64-67 134-156 | Yield (%) SO ₄ ²⁻ group den- sity (mmol/kg) | 66-69 241-265 CCD minimize variables limitations [85] |
| Orthogonal | Microcrystalline cellulose | Acid conc. (wt.%) Temp. (°C) Time (min) | 33 43 108 | Yield (%) 90.14 | Reaction time affect the yield most [87] |

when a massive number of variables are involved, it will become more tedious and inefficient as numerous experiments will have to be conducted to determine the optimum point. In fact, there are chances that the optimum level will be missed out during the analysis [89].

For this reason, it is essential to use optimization tools when conducting an experimental process that consists of multi-level parameters. By implementing optimization, especially in the synthesis part, the overall operational cost can be minimized, thereby maximizing the production efficiency. Adapting an experimental design to meet the needs of a process will require a well-fitted model in relation to the experimental data obtained. As optimization can evaluate the correlation between each factor and dependent outcomes with a minimum number of experiments, it can increase productivity and efficiency, particularly on an industrial scale.

Conclusion

The growing scientific and industrial interest in nanomaterials has led to the development of various methods for the preparation of starch-based nanoparticles. Currently, the implementation of acid-treated starch is gaining tremendous attention globally due to its unique properties and outstanding performance in many applications. The fascinating characteristics of nanostarch depend on the hydrolysis conditions, namely, the acid, concentration of acid, reaction time, reaction temperature, and source of starch. Throughout this review, it was recommended that organic acid be used instead of inorganic acids in the preparation of nanostarch because organic acid is more selective, effective, environmentally friendly, as well as non-corrosive to the process equipment. Furthermore, organic acid possesses appealing practical and chemical characteristics, such as recoverability, and ease of handling and storage. This current review also revealed that the recovery yield and particle size of the nanostarch are vital in defining the optimum value of each of the process conditions. Therefore, researchers have discovered an improved technique for the preparation of nanostarch with the desired properties and maximum yield by adapting the optimization process. The use of RSM to optimize the starch modification process is now being widely used and consolidated owing to its advantages over the traditional one-factor-at-a-time optimization, such as the ability to generate large amounts of data from a small number of experiments, and the ability to evaluate the interaction effects between multiple variables on the response surface. In this current review, it is suggested that a high yield of nanostarch with the desired properties can be prepared by optimizing the acid hydrolysis conditions with a statistical experimental design and RSM.

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